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VOLUME 4

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High Altitude Sampling Program

THE APPLICATION OF HASP DATA

ISOTOPES, INC.

Defense Atomic Support Agency

WASHINGTON 25, D.C.

DASA 1300

THE HIGH ALTITUDE SAMPLING PROGRAM

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VOLUME 4

THE APPLICATION OF HASP DATA

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CHAPTER 7

THE SURFACE BURDEN OF WORLD-WIDE FALLOUT

One of the major objectives of the High Altitude Sampling Program is to assess the rate at which radioactive debris from world-wide fallout is deposited on the earth's surface following the detonations of high yield nuclear weapons. Much of the debris from these detonations is injected into the stratosphere and the time required for its subsequent deposition is relatively long, several months to several years, compared to the few weeks required for the deposition of tropospheric debris. The most direct approach to the determination of the surface burden of nuclear fallout involves the measurement of the concentrations of radionuclides on the land and in the oceans. Alternatively the concentrations of radioactivity may be measured in samples of precipitation and these data may be integrated to give the total deposition of nuclear debris during the time interval of interest. Such studies yield valuable information on the mechanisms involved in the deposition of fallout and in addition they permit estimates of the present and future radiological hazards to be made. In this chapter we are concerned primarily with past and future surface burdens of strontium-90 and tungsten-185 and their variations with time.

Several workers have approached the problem of the calculation of the surface burden of world-wide fallout in different ways. A total of five alternative methods has been proposed, four of which include the use of precipitation data in conjunction with the results of analyses of soils for radioactive fallout. Since radioactive fallout is deposited on the earth's surface primarily by precipitation, it is believed that these four approaches yield more accurate estimates of the total surface burden. All five methods are outlined and discussed in this chapter.

Recently, data on the strontium-90 contents of soils collected in 1959 from a world-wide network of stations were published[1]. Calculations, described in this chapter, of the surface burden of strontium-90 for mid-1959 were performed using these data and extrapolations of the burden to 1960 and future years were carried out using present theories on the mechanisms of fallout and information on current concentrations of fission products in precipitation. Based on similar procedures, computations of the surface burden of strontium-90 for previous years are reviewed to enable estimates of the annual increments to be made.

In assessing the distribution of strontium-90 on the surface of the earth consideration must be given to the contribution of activity from the Nevada test series. A new approach to the calculation of the intermediate fallout fraction from this source was proposed recently[2] and is reviewed in this chapter.

Since mid-1958 tungsten-185 has been determined in rain water samples at several stations throughout the world. This nuclide was produced during the Hardtack test series in mid-1958 and its subsequent history in the stratosphere has contributed substantially to our knowledge of the movement of nuclear debris in the stratosphere[3]. The pattern of its deposition on the ground is also of extreme importance to our understanding of the mechanisms of stratospheric fallout and a review is given in this chapter of the rate and distribution of fallout of tungsten-185 from the time of its release until the end of 1959, when it was no longer detectable in precipitation. This information may be used, in conjunction with stratospheric data to clarify the details of its injection into the stratosphere, its mixing and transfer within the stratosphere and troposphere and its final deposition on the surface of the earth.

COMPUTATION OF SURFACE BURDEN OF STRONTIUM-90

It was indicated above that five methods have been suggested for the computation of the total amount of strontium-90 on the earth's surface at a given time. Two of these procedures, developed in HASP, are presented in detail. The three remaining approaches are described briefly. It is worthwhile mentioning at this point that none of these methods takes into account direct measurements of the amounts of fallout in the oceans. In all procedures it is assumed that measurements made on land can be extrapolated over the oceans without regard to possible differences in fallout mechanisms which may exist.

HASP Calculations--Method 1

HASP calculations of the world-wide deposit of strontium-90 are based on the approximation formula:

$$F = \sum_{i=1}^{i=N} A_i \bar{f}_i \bar{p} \qquad (1)$$

where F = total deposit of strontium-90 on the earth's surface (in mc).

A_i = area (mi^2) of latitude belt i.

$\bar{f}_i$ = mean "deposition concentration" of strontium-90 in the latitude belt i, expressed in mc/mi^2/inch of mean annual rainfall (MAR). This quantity is calculated for each soil sampling station from the strontium-90 deposit (mc/mi^2) and the MAR at the site.

$\bar{p}$ = mean annual rainfall (MAR) in inches, for the latitude belt i (Möller's[4] values are accepted).

Equation (1) assumes that there is no correlation between f and p, i.e., the deposition concentration of strontium-90 is independent of the amount of precipitation. While this is true in certain restricted climatic regimes, such as in the United Kingdom it may not be absolutely correct when larger areas encompassing several different climatic regimes are considered. Further discussion of the possible relationship between f and p is presented in Methods 3 and 4 which follow.

Results of the calculations of f_i for the data on strontium-90 in soils collected in 1959 are shown in Table 7.1. All deposition concentrations were corrected to a reference date of July 1, 1959 and the complete procedure used in the calculation of the surface burden of strontium-90 is as follows:

1. All the reported soil activities were corrected to July 1, 1959. The increment to be added or subtracted from the measured strontium-90 deposit (mc/mi^2) was determined from the strontium-90 concentration in precipitation for the period of correction (Tables 7.2 and 7.3) and the rainfall during the period.

2. Because all soil samples were taken to 6 inches depth or greater it was assumed that 100% of the strontium-90 deposit at the site was obtained and measured in each sample.

3. The cumulative deposit (mc/mi^2) at each site on July 1, 1959 was divided by the MAR, computed from the measured precipitation at the site during the period from January 1953 to the time of sampling, to obtain the deposition concentration.

4. An average deposition concentration (mc/mi^2/inch MAR) of strontium-90 was calculated for each 10° latitude band by averaging the soil results for all sites within the band.

5. The total amount of strontium-90 in each band was obtained by multiplication of the average deposition concentration in the band by the MAR for the band (calculated from Möller's data[4]) and by the area of the band.

6. The total surface burden was then calculated by summation of the deposits in each latitude band.

7. Sample sites falling within the intermediate fallout zone around the Nevada Test Site, as shown in a following section, were omitted from this calculation.

The results of the above calculations are shown in Tables 7.4 and 7.5. In Table 7.4 are given the computed average strontium-90 concentrations in each 10° latitude band and the idealized concentrations which have been used in the final calculation of the surface burden of strontium-90. The idealized concentrations were derived by assuming that the deposition concentrations are constant throughout the 30° - 90° zone in each hemisphere. The deposition

concentration in a 30° - 90° zone is determined by weighting the average concentration in each of the included 10° latitude zones by the number of sampling stations within it and computing the average of these combined observations.

Table 7.5 summarizes the final calculation of the surface burden of strontium-90 from soil analyses for July 1, 1959. A deposit of 3.63 megacuries is calculated for this date.

HASP Calculations--Method 2

A second approach used in HASP studies for the calculation of the surface burden of strontium-90 is based entirely on the data obtained from worldwide observations of strontium-90 concentrations in precipitation. Data such as those shown in Table 7.2 are available[5] from 1954 through June 1960. From these results the average strontium-90 concentration in precipitation was calculated for each twelve month period for each 10° latitude band. Multiplication of the average annual strontium-90 concentration in precipitation for the latitude band by the mean annual rainfall (MAR) for the band and by the area of the band yielded the increment of strontium-90 deposited during the previous twelve month period. Summation of the annual deposits of strontium-90 from 1954 to 1960 gave the total amount of strontium-90 deposited on the surface of the earth up to July 1, 1960. In this method it is obvious that radioactive decay of strontium-90 is not taken into account. Hence, the surface burdens of strontium-90 obtained by this procedure can yield higher results than those calculated according to Method 1. As in the procedures described in Method 1 some approximations were made for the areas where data were sparse. Thus, idealized concentrations were calculated both for 30°N to 90°N and 30°S to 90°S in the same manner as previously described. These values are shown in Table 7.6. From these results total strontium-90 deposits of 3.89 and 4.47 megacuries on the earth's surface on July 1, 1959 and July 1, 1960, respectively, were calculated.

Table 7.1. Strontium-90 Deposition in Soils--1959

Station	Latitude (degrees)	Collection Date	Measured Surface Deposit (mc Sr^{90}/mi^2)	Total* Rainfall (inches)	MAR ** (inches)	Surface Deposit (mc Sr^{90}/mi^2) on 7-1-59	Deposition Concentration (mc Sr^{90}/mi^2/ inch MAR)
70° - 80°N							
Barrow, Alaska	71.3	6-24	9.9	33	5	10.0	2.00
Barrow, Alaska	71.3	6-24	12.3	33	5	12.4	2.48
Vadso, Norway	70.1	6-25	17.8	?	?	---	---
Ekkeroy, Norway	70.1	6-25	23.7	113	17.4	24.1	1.38
60° - 70°N							
Bardufoss, Norway	69.2	6-23	35.4	174	26.9	36.0	1.34
Aklavik, Canada	68.2	7-23	5.1	47	7.1	5.0	0.70
Bodo, Norway	67.3	6-23	51.4	268	41.3	52.4	1.27
Fairbanks, Alaska	64.8	6-23	19.1	61	9.3	19.3	2.08
Reykjavik, Iceland	64.1	6-26	64.0	221	34.1	64.5	1.89
Ft. Simpson, Canada	61.9	6-23	15.3	82	12.7	15.6	1.23
Palmer, Alaska	61.6	6-19	22.0	90	13.9	22.5	1.62
Bergen, Norway	60.4	6-19	83.8	503	77.9	86.8	1.11
50° - 60°N							
Oslo, Norway	59.9	6-18	41.2	174	26.9	42.3	1.57
Ft. Chimo, Canada	58.1	6-17	11.4	105	16.3	12.2	0.75
Lacombe, Canada	53.0	9-25	39.3	126	18.6	38.3	2.06
40° - 50°N							
Courtenay, Canada	49.7	9-22	78.5	395	58.6	7 .2	1.28
Agassiz, Canada	49.2	9-21	90.1	466	59.0	8b 3	1.25
Douglas, Canada	49.0	9-22	73.2	260	38.5	71.0	1.84
Paris, France	49.0	6-17	35.4	116	17.9	36 2	2.02
Saanichton, Canada	48.6	9-23	44.3	230	34.1	42.4	1.24
Clallam Bay, Wash.	48.3	9-24	71.2	566	83.9	66.4	0.79
Joyce, Wash.	48.1	9-24	48.7	404	59.8	45.4	0.76
Pt. Angeles, Wash.	48.1	9-24	57.8	187	27.7	56.3	2.03
Seguim, Wash.	48.1	9-24	29.7	119	17.6	28.8	1.64
Forks, Wash.	48.0	9-25	99.6	812	120.5	93.0	0.77
St. Johns, Nfd.	47.5	9-30	72.3	428	63.5	68.5	1.08
Payallup, Wash.	47.2	9-21	54.5	289	42.8	52.1	1.22
Mandan, N. D.	46.8	10-21	46.1	115	16.9	45.0	2.66+
Longview, Wash.	46.2	9-25	61.5	322	47.7	58.9	1.24
Bozeman, Mont.	45.7	10-19	73.9	112	16.5	72.8	4.41+
Ottawa, Canada	45.4	6-17	50.7	204	31.4	52.2	1.66
Kentville, N. Scotia	45.1	6-24	47.3	285	43.9	48.3	1.10
St. Paul, Minn.	45.0	10-21	59.1	170	24.9	57.5	2.31+
Orono, Maine	44.9	8-17	49.3	240	35.9	48.1	1.34
Corvallis, Oregon	44.6	9-27	57.5	291	42.8	55.0	1.28
Burlington, Vermont	44.5	8-18	50.9	228	34.4	49.8	1.45

+ Believed to be influenced by intermediate fallout from Nevada tests.

Table 7.1 (continued)

Station	Latitude (degrees)	Collection Date	Measured Surface Deposit (mc Sr^{90}/mi^2)	Total* Rainfall (inches)	MAR** (inches)	Surface Deposit (mc Sr^{90}/mi^2) on 7-1-59	Deposition Concentration (mc Sr^{90}/mi^2/ inch MAR)
40° - 50°N (continued)							
Rapid City, S. D.	44.1	4-25	78.6	111	17.3	82.4	4.76+
Florence, Italy	43.8	6-15	52.2	183	28.3	53.6	1.89
Boise, Idaho	43.6	10-15	56.6	82	11.9	55.8	4.69+
Madison, Wis.	43.1	10-22	60.7	206	30.0	58.7	1.96
Durham, N. H.	43.1	8-16	57.5	292	43.4	56.2	1.29
Rochester, N. Y.	43.1	10-7	59.3	215	31.5	57.4	1.82
Sapporo, Japan	43.0	6-5	58.2	305	47.6	62.2	1.31
Glendale, Oregon	42.8	9-27	50.6	303	44.5	48.0	1.08
Ithaca, N. Y.	42.4	10-7	51.7	244	35.6	49.5	1.39
Amherst, Mass.	42.4	8-19	52.1	291	43.1	50.7	1.18
Detroit, Mich.	42.2	11-3	52.3	200	28.9	49.5	1.71
Binghamton, N. Y.	42.2	10-8	55.0	251	37.2	52.6	1.41
S. Wellfleet, Mass.	42.1	8-15	82.5	274	40.9	81.3	1.99
Windsor, Conn.	41.9	8-14	59.6	294	43.9	58.3	1.33
Logan, Utah	41.8	10-14	45.3	102	14.8	44.3	2.99+
Des Moines, Iowa	41.5	10-20	65.6	183	26.5	63.9	2.41+
Kingston, R. I.	40.5	8-14	99.2	329	49.0	97.8	2.00
Lincoln, Neb.	40.8	5-25	56.6	178	27.8	60.1	2.16+
New York, N. Y.	40.8	8-13	65.6	263	39.2	64.5	1.65
Penn. Univ., Penn.	40.8	8-26	66.7	262	39.0	65.3	1.67
Salt Lake City, Utah	40.8	10-13	100.6	113	16.4	99.5	6.06+
New Brunswick, N. J.	40.5	10-8	62.5	288	42.4	59.9	1.41
W. Lafayette, Ind.	40.4	9-2	77.3	238	35.2	75.9	2.16
Columbus, Ohio	40.0	9-1	68.1	222	32.9	66.8	2.03
30° - 40°N							
Springfield, Ill.	39.8	11-4	67.2	222	32.1	65.0	2.02
Derby, Colo.	39.8	4-24	54.6	99	15.5	58.1	3.74+
Newark, Del.	39.7	8-13	63.9	284	42.3	62.7	1.48
Morgantown, W. Vir.	39.6	8-31	74.0	261	38.6	72.4	1.88
Manhattan, Kan.	39.2	11-17	76.9	201	29.0	74.8	2.58+
Columbia, Mo.	39.0	11-16	86.0	222	31.8	83.7	2.63+
Healdsburg, Cal.	38.6	9-29	55.3	285	41.3	52.8	1.28
Lexington, Ky.	38.0	9-2	74.9	292	43.3	73.2	1.69
Blacksburg, Vir.	37.2	9-4	55.7	255	37.8	54.1	1.43
Tulsa, Okla.	36.2	11-5	74.0	239	34.6	71.5	2.06
Knoxsville, Tenn.	35.8	9-3	67.3	301	44.6	65.5	1.47
Raleigh, N. C.	35.8	11-9	50.6	343	49.5	47.1	0.95
Tokyo, Japan	33.7	6-4	65.5	409	64.0	71.1	1.11
Little Rock, Ark.	34.7	11-5	65.6	346	50.0	62.1	1.24
Clemson, S. C.	34.7	11-10	63.5	345	49.8	60.0	1.21
Los Angeles, Cal.	33.9	12-29	23.6	74	10.6	22.7	2.14
Atlanta, Ga.	33.6	11-10	60.2	301	43.4	57.1	1.32
Fukuoka, Japan	33.6	6-8	48.0	490	75.4	53.6	0.71
Birmingham, Ala.	33.6	12-21	72.8	344	49.2	70.8	1.44
Brawley, Cal.	33.0	1-27	7.7	7.4	1.2	7.8	6.49+
El Centro, Cal.	32.8	12-31	15.5	15	2.1	15.3	7.29+
Newton, Miss.	32.3	12-21	71.1	361	51.6	66.8	1.30
Hamilton, Bermuda	32.3	9-14	69.5	444	65.5	66.2	1.01
Tucson, Arizona	32.2	12-25	30.8	72	10.0	30.0	3.00+
Waco, Texas	31.6	12-23	61.4	208	29.8	58.9	1.98
Alapaha, Ga	31.4	11-11	50.0	313	45.1	47.2	1.05
Alexandria, La.	31.3	12-22	59.1	379	54.2	54.8	1.01
Jacksonville, Fla.	30.4	11-12	62.8	356	51.2	59.1	1.15

+ Believed to be influenced by intermediate fallout from Nevada tests.

Table 7.1 (continued)

Station	Latitude (degrees)	Collection Date	Measured Surface Deposit (mc Sr^{90}/mi^2)	Total* Rainfall (inches)	MAR** (inches)	Surface Deposit (mc Sr^{90}/mi^2) on 7-1-59	Deposition Concentration (mc Sr^{90}/mi^2/ inch MAR)
20° - 30°N							
S. Miami, Fla.	25.8	7-6	48.6	410	62.1	48.4	0.78
Oahu, Hawaii 1	21.7	6-1	52.0	273	42.0	53.1	1.26
2	21.5	6-2	120.0	1204	185.0	124.7	0.67
3	21.5	6-1	57.1	364	56.1	58.6	1.04
4	21.4	6-1	58.9	406	62.4	60.5	0.97
Maui, Hawaii 1	20.9	9-29	388	2506	363	375.4	1.03
2	20.8	8-31	285	2078	308	277.7	0.90
10° - 20°N							
San Juan, Puerto Rico	18.5	7-27	51.8	393	59.6	51.6	0.87
Manila, Philipines	14.6	6-10	13.0	445	69.0	14.0	0.20
0° - 10°N							
Ft. Clayton, Panama	9.0	7-7	18.8	501	76.8	18.7	0.24
Ft. Amador, Panama	9.0	7-7	16.1	473	72.6	16.1	0.22
Singapore	1.4	6-12	9.0	573	89.0	9.1	0.10
0° - 10°S							
Kikuyu, Kenya	1.2	7-6	9.8	230	35.3	9.8	0.28
Belem, Brazil	1.5	7-10	18.3	706	108.0	18.2	0.17
10° - 20°S							
Huancayo, Peru	12.0	6-11	8.8	191	29.6	8.9	0.30
Katherine, Australia	14.3	6-12	7.7	258	40.0	7.8	0.20
Salisbury, S. Rhodesia	17.8	7-1	6.5	235	36.2	6.5	0.18
20° - 30°S							
Alice Springs, Australia	23.8	6-12	4.9	62	9.6	4.9	0.51
Brisbane, Australia	27.5	6-12	13.9	302	46.9	14.1	0.30
30° - 40°S							
Perth, Australia	32.0	6-12	6.9	222	32.0	7.0	0.22
Adelaide, Australia	34.9	6-12	13.0	125	19.4	13.1	0.67
N. Auckland, N. Z.	35.8	6-16	14.4	470	72.8	14.7	0.20
40° - 50°S							
Wellington, N. Z.	41.3	6-15	15.8	338	52.4	16.0	0.31
Hobart, Tasmania	42.9	6-12	15.4	189	29.3	15.5	0.53
S. Canterbury, N. Z.	44.4	6-17	10.8	148	22.9	10.9	0.48
50° - 60°S							
Punta Arenas, Chile	53.2	10-	7.9	121	17.8	7.7	0.43
Campbell Island	52.5	6-15	15.2	369	57.1	15.4	0.27

* Total rainfall as observed at, or close to, the soil sampling station from 1-1-53 to collection date of soil sample.

** MAR - Mean Annual Rainfall calculated from total rainfall at the site.

Table 7.2. Concentrations of Strontium-90 in Rain Water During 1959

Station	Latitude (degrees)	Average Quarterly Concentration (mc $Sr^{90}/mi^2/inch$)			
		1	2	3	4
60° - 70°N					
Tromso, Norway	70	1.61	---	---	---
Bodo, Norway	67	1.37	---	---	---
Oslo, Norway	60.5	---	---	0.52	0.07
50° - 60°N					
Kinloss, Scotland	58	0.62	1.17		---
Prestwick, Scotland	57	---	---		0.06
Liverpool, England	54	0.74	1.24		---
Snowdow, England	53	0.78	1.25		---
Felixstowe, England	52	0.98	1.46		---
Abingdon, England	51	0.72	1.22		---
Milford Haven, England	51	0.88	1.34		---
Sylt, Germany	51	---	1.62		---
40° - 50°N					
Williston, N. D.	49	0.43	1.59	0.08	0.07
International Falls, Minn.	48	0.29	1.25	0.36	0.07
Seattle, Wash.	48	0.70	1.43	0.31	0.06
Vienna, Austria	48	0.66	0.93	0.32	0.04
Ottawa, Canada	48	0.91	---	---	---
Helena, Mont.	47	0.51	1.29	0.71	0.13
Klagenfurt, Austria	47	0.38	0.58	0.27	0.05
Milan, Italy	45	---	---	0.01	0.01
Vermillion, S. D.	44	1.47	1.18	0.16	0.13
Columbus, Wis.	44	0.49	---	---	---
Green Bay, Wis.	44	0.32	1.11	0.11	0.08
Florence, Italy	43	---	---	---	0.05
Medford, Ore.	42	0.33	2.03	0.53	0.10
Salt Lake City, Utah	41	2.52	2.76	0.66	1.00
Lemont, Ill.	41	0.47	0.67	0.18	---
Pittsburgh, Penn.	41	0.74	1.01	0.18	0.08
New York City, N. Y.	41	0.75	1.07	0.13	0.08
Westwood, N. J.	41	0.87	1.33	0.20	0.07
30° - 40°N					
Columbia, Mo.	39	1.37	1.04	0.04	0.11
Denver, Colo.	40	0.48	2.43	0.38	0.10
Lajes Field, Azores	38	---	---	---	0.03
Louisville, Kentucky	38	0.52	0.45	0.09	0.09
Richmond, Cal.	38	0.30	0.70	0.02	0.05
San Francisco, Cal.	37	0.11	1.83	0.04	---
Tulsa, Okla.	37	1.93	0.94	0.12	0.07
Gibraltar	36	1.17	---	---	---
Hiroshima, Japan	36	0.96	0.48	0.01	0.05
Columbia, S. C.	34	---	0.39	0.38	0.06
Nagasaki, Japan	34	0.70	0.55	0.01	0.06
W. Los Angeles, Cal.	34	1.08	1.62	0.68	---
Birmingham, Ala.	33	0.70	0.45	0.22	0.03
Dallas, Texas	33	0.64	1.29	0.07	0.01
El Paso, Texas	33	---	2.43	0.30	0.19
Beirut, Lebanon	32	---	---	---	0.05
20° - 30°N					
Houston, Texas	29	0.36	0.53	0.08	0.03
Coral Gables, Fla.	27	0.87	0.30	0.09	0.02
Karachi, Pakistan	26	---	---	0.04	---
Taipei, Taiwan	24	0.66	0.16	0.02	0.09
Itilo, Hawaii	22	0.46	0.27	0.14	0.01
Oahu, Hawaii	22	1.00	0.70	0.73	0.15

Table 7.2 (continued)

Station	Latitude (degrees)	Average Quarterly Concentration (mc Sr^{90}/mi^2/inch) 1	2	3	4
10° - 20°N					
Caenwood, Jamaica	18	0.55	---	---	---
Palisadoes, Jamaica	18	0.89	---	---	---
Bangkok, Thailand	14	0.27	0.03	0.03	0.01
Wake Island	19	---	0.48	0.06	0.07
Mauno Loa, Hawaii	19	0.36	0.03	0.02	0.35
0° - 10°N					
Turrialba, Costa Rica	10	---	---	0.01	---
Canal Zone	9	---	---	0.02	0.01
Monrovia, Liberia	6	---	---	0.05	0.02
Lagos, Nigeria	6	0.24	---	0.02	0.01
Singapore	1	0.07	0.03	---	---
0° - 10°S					
Belem, Brazil	2	---	---	---	0.01
Nairobi, Kenya	2	---	---	0.07	0.02
Kikuyu, Kenya	2	0.11	0.08	0.05	0.01
Mohaus, Brazil	3	0.09	0.01	0.02	0.02
Canton Island	3	0.05	0.01	0.01	0.10
10° - 20°S					
Lima, Peru	16	---	---	---	0.04
Darwin, Australia	17	0.05	0.01	0.05	0.02
Suava, Fiji	18	≤ 0.20	---	0.02	0.01
Salisbury, S. Rhodesia	18	0.04	0.07	0.75	0.02
Townsville, Australia	19	0.04	0.03	---	0.01
20° - 30°S					
Pretoria, S. Africa	25	0.10	0.11	0.22	0.05
Brisbane, Australia	27	0.05	0.10	0.07	0.07
Durban, S. Africa	29	0.09	0.06	0.03	0.11
30° - 40°S					
Perth, Australia	32	0.42	0.06	0.07	0.08
Santiago, Chile	33	0.01	0.08	0.12	0.14
Adelaide, Australia	35	0.17	0.19	0.12	0.09
Sydney, Australia	35	0.08	0.09	0.04	0.07
Melbourne, Australia	38	0.12	0.09	0.06	0.12
40° - 50°S					
Ohakea, New Zealand	40	0.11	---	---	---
Lower Hutte, New Zealand	41	0.04	0.01	0.02	0.01
Wellington, New Zealand	41	0.16	---	---	0.04
Hobart, Tasmania	42	0.19	0.08	0.07	0.58
50° - 60°S					
Port Stanley, Falkland Is.	52	0.11	---	---	---

Table 7.3. Idealized Quarterly Concentrations ($mc/mi^2/in$) of Strontium-90 in Rain Water During the Second Half of 1958 and During 1959

Latitude Band	1958		1959			
	3	4	1	2	3	4
30° - 90°N	0.30	0.35	0.82	1.24	0.24	0.10
20° - 30°N	0.16	0.16	0.62	0.33	0.14	0.10
10° - 20°N	0.07	0.07	0.57	0.26	0.05	0.04
0° - 10°N	0.03	0.03	0.16	0.03	0.03	0.01
0° - 10°S	0.03	0.03	0.08	0.04	0.04	0.05
10° - 20°S	0.03	0.03	0.03	0.09	0.04	0.02
20° - 30°S	0.10	0.10	0.08	0.10	0.10	0.08
30° - 90°S	0.07	0.11	0.14	0.09	0.07	0.14

Table 7.4. Average Strontium-90 Deposition Concentrations in Soil on July 1, 1959

Latitude Band	Soil Stations Within Band	Deposition Concentration ($mc/mi^2/in.$ MAR)	
		Computed	Idealized
80° - 90°N	0	---	1.47
70° - 80°N	3	1.95	1.47
60° - 70°N	8	1.40	1.47
50° - 60°N	3	1.46	1.47
40° - 50°N	36	1.48	1.47
30° - 40°N	22	1.41	1.47
20° - 30°N	7	0.95	0.95
10° - 20°N	2	0.54	0.54
0° - 10°N	3	0.19	0.19
0° - 10°S	2	0.22	0.22
10° - 20°S	3	0.23	0.23
20° - 30°S	2	0.40	0.40
30° - 40°S	3	0.36	0.39
40° - 50°S	3	0.43	0.39
50° - 60°S	2	0.35	0.39
60° - 70°S	0	---	0.39
70° - 80°S	0	---	0.39
80° - 90°S	0	---	0.39

Table 7.5. Surface Burden* of Strontium-90 on 1 July 1959

Latitude Band	Area ($mi^2 \times 10^{-6}$)	MAR** (inches)	Deposition Concentration (mc/mi^2/in. MAR)	Deposit (megacuries)
80° - 90°N	1.6	4	1.47	0.009
70° - 80°N	4.3	6	1.47	0.038
60° - 70°N	7.3	14	1.47	0.150
50° - 60°N	9.8	26	1.47	0.375
40° - 50°N	12.2	31	1.47	0.556
30° - 40°N	14.0	28	1.47	0.576
20° - 30°N	15.5	25	0.95	0.378
10° - 20°N	16.5	34	0.54	0.303
0° - 10°N	17.1	61	0.19	0.198
0° - 10°S	17.1	46	0.22	0.173
10° - 20°S	16.5	37	0.23	0.140
20° - 30°S	15.5	26	0.40	0.161
30° - 40°S	14.0	31	0.39	0.169
40° - 50°S	12.2	44	0.39	0.209
50° - 60°S	9.8	38	0.39	0.145
60° - 70°S	7.3	16	0.39	0.046
70° - 80°S	4.3	3	0.39	0.005
80° - 90°S	1.6	1	0.39	0.001
				3.63 MC

* Calculated from soil data.

** Mean Annual Rainfall from Möller.

Table 7.6. Idealized Concentrations (mc Sr^{90}/mi^2/inch) of Strontium-90 in Precipitation for the Period 1954 - 1959

Quarter	South Latitude				North Latitude			
	90°-30°	30°-20°	20°-10°	10°-0°	0°-10°	10°-20°	20°-30°	30°-90°
1954								
1	0.03	0.01	0.01	0.01	0.01	0.02	0.06	0.11
2	0.03	0.01	0.01	0.01	0.01	0.03	0.11	0.16
3	0.05	0.03	0.01	0.01	0.01	0.03	0.06	0.09
4	0.05	0.03	0.02	0.02	0.02	0.03	0.08	0.10
1955								
1	0.06	0.02	0.02	0.02	0.02	0.04	0.13	0.23
2	0.06	0.06	0.02	0.02	0.03	0.09	0.24	0.36
3	0.07	0.12	0.03	0.03	0.03	0.07	0.16	0.24
4	0.07	0.12	0.01	0.01	0.02	0.04	0.08	0.10
1956								
1	0.07	0.03	0.02	0.02	0.02	0.06	0.19	0.33
2	0.07	0.06	0.02	0.02	0.03	0.12	0.32	0.47
3	0.09	0.06	0.02	0.02	0.03	0.05	0.12	0.18
4	0.09	0.05	0.02	0.02	0.03	0.06	0.14	0.19
1957								
1	0.09	0.02	0.02	0.02	0.02	0.05	0.14	0.24
2	0.09	0.04	0.03	0.03	0.03	0.08	0.20	0.30
3	0.06	0.06	0.03	0.03	0.02	0.05	0.13	0.20
4	0.09	0.06	0.03	0.03	0.02	0.05	0.12	0.16
1958								
1	0.07	0.02	0.02	0.02	0.02	0.05	0.18	0.25
2	0.04	0.04	0.03	0.03	0.03	0.09	0.28	0.58
3	0.07	0.10	0.03	0.03	0.03	0.07	0.16	0.30
4	0.11	0.10	0.03	0.03	0.03	0.07	0.16	0.35
1959								
1	0.14	0.08	0.03	0.08	0.16	0.57	0.62	0.82
2	0.09	0.10	0.09	0.04	0.03	0.26	0.33	1.24
3	0.07	0.10	0.04	0.04	0.03	0.05	0.14	0.24
4	0.14	0.08	0.02	0.05	0.01	0.04	0.10	0.10
1960								
1	0.09	0.04	0.04	0.04	0.06	0.17	0.15	0.21
2	0.04	0.05	0.06	0.03	0.02	0.20	0.10	0.24

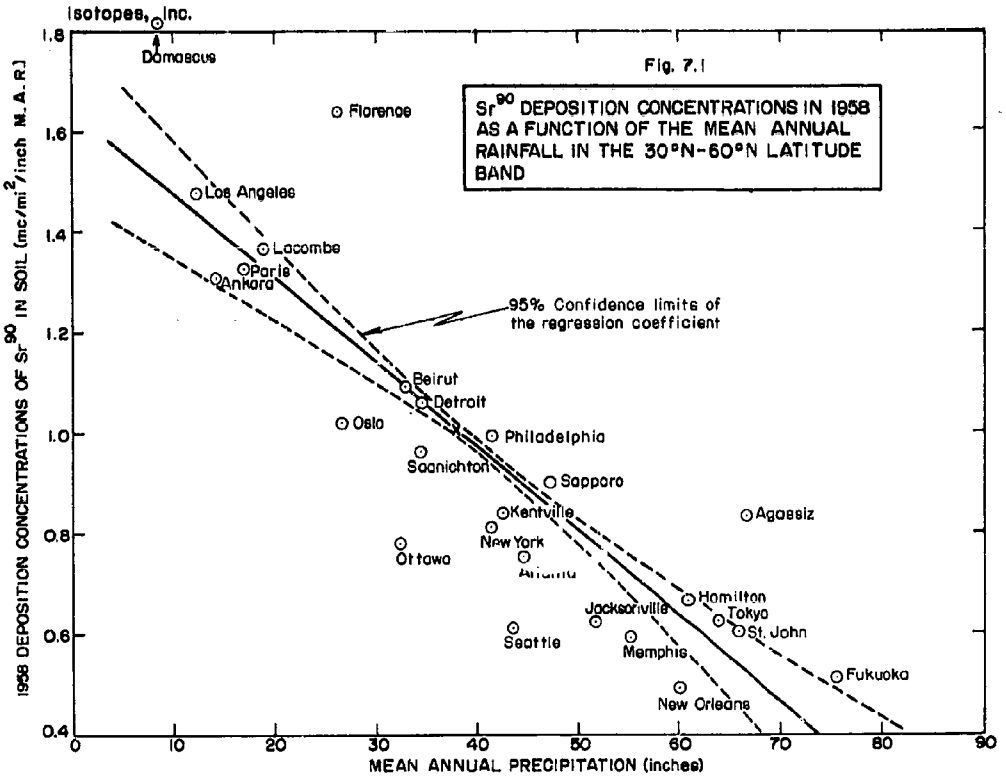
Alternative Methods

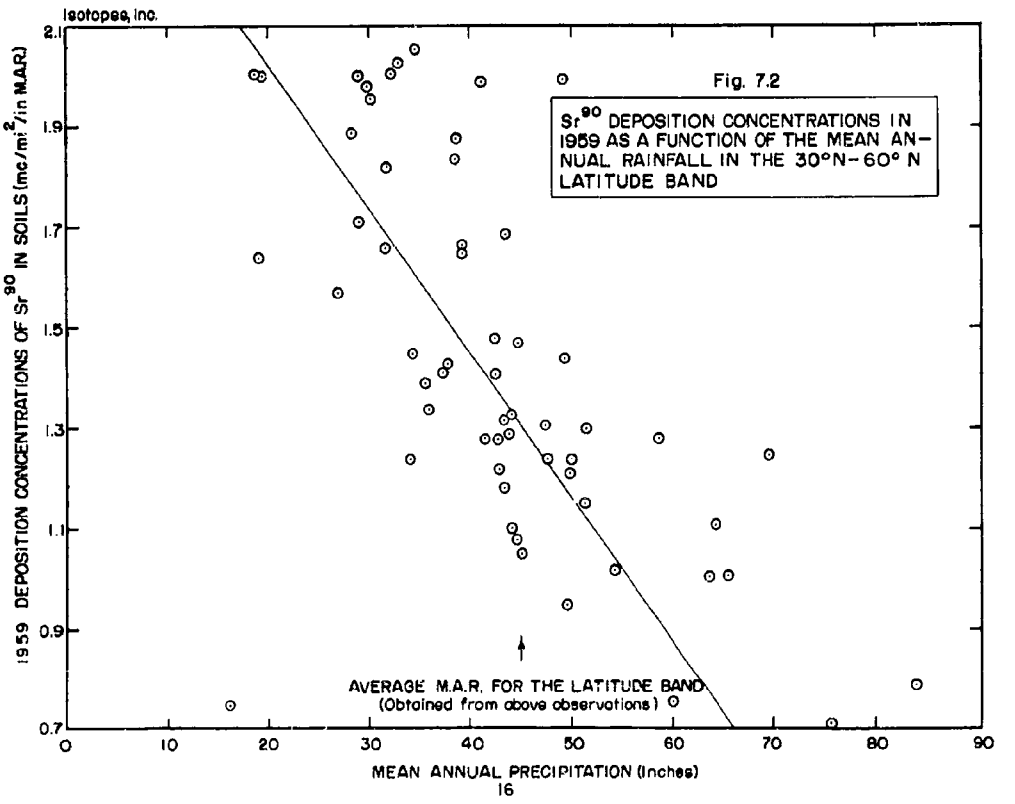
Three alternative methods have been proposed for the calculation of the deposit of strontium-90 on the earth's surface. Two of these methods stem from the observation that in certain latitude bands it appears that a relationship exists between the amount of precipitation and the deposition concentration of strontium-90[1,6]. In general it was observed that areas with varying amounts of precipitation but within the same climatic regime exhibited approximately the same deposition concentration of strontium-90. However, when different climatic regimes from within the same latitude zone were examined it was apparent that high rainfall areas possessed lower deposition concentrations of strontium-90 than low rainfall regions. In these two methods of calculation of the surface burden of strontium-90 an attempt was made to take this observation into consideration.

Method 3

This method consists of computation of the deposition concentrations of strontium-90 for several sampling sites within a latitude band and then determination of an empirical relationship between the deposition concentration at each site and the measured mean annual rainfall at each site. The mean deposition concentration of strontium-90 for the latitude band is then determined by that deposition concentration corresponding to the MAR for the whole latitude band. Multiplication of the mean deposition concentration for a latitude band by the MAR and by the area of that band yields the strontium-90 deposit within the latitude band. Summation of the zonal deposits gives the total world-wide strontium-90 surface burden.

The empirical relationship between the deposition concentration of strontium-90 and the MAR (for a single site) within a latitude band is illustrated in Figures 7.1 and 7.2. Here (for 1958 and 1959 respectively) each


Isotopes, Inc.
Damascus
Fig. 7.1
Sr90 DEPOSITION CONCENTRATIONS IN 1958 AS A FUNCTION OF THE MEAN ANNUAL RAINFALL IN THE 30°N-60°N LATITUDE BAND
Florence
Los Angeles
Lacombe
Paris
Ankara
95% Confidence limits of the regression coefficient
Beirut
Detroit
Oslo
Philadelphia
Saanichton
Sapporo
Kentville
Agassiz
New York
Ottawa
Hamilton
Jacksonville
Tokyo
St. John
Seattle
Memphis
Fukuoka
New Orleans
1958 DEPOSITION CONCENTRATIONS OF Sr90 IN SOIL (mc/mi2/inch M.A.R.)
MEAN ANNUAL PRECIPITATION (inches)


Isotopes, Inc.
Fig. 7.2
Sr90 DEPOSITION CONCENTRATIONS IN 1959 AS A FUNCTION OF THE MEAN ANNUAL RAINFALL IN THE 30°N-60° N LATITUDE BAND
AVERAGE M.A.R. FOR THE LATITUDE BAND
(Obtained from above observations)
1959 DEPOSITION CONCENTRATIONS OF Sr90 IN SOILS (mc/mi2/in M.A.R.)
MEAN ANNUAL PRECIPITATION (Inches)

point represents a single site within the 30°N - 60°N latitude band. The deposition concentration for each location was determined from the soil analysis result (mc/mi^2) and the mean annual rainfall calculated from the total amount of precipitation at the site from January 1953 until the time of sampling. From the scatter of points it is clear that there exists an approximately linear relationship (with a negative slope) between strontium-90 deposition concentration and MAR.

The MAR for the 30°N - 60°N latitude band given by Möller (based on more comprehensive precipitation data than are at our disposal from the individual soil sampling studies) is ~27 inches. From Figure 7.2 it is found that corresponding to Möller's estimate of MAR for the 30°N - 60°N latitude zone is a deposition concentration of strontium-90 of 1.82 mc/mi^2/inch MAR, which is assumed to be the mean deposition concentration for the whole latitude band.

To obtain the total deposit of strontium-90 on the surface of the earth, the above procedure was applied to other latitude zones. Similar curves of strontium-90 deposition concentration as a function of MAR were plotted for the following zones; 20°N - 30°N, 10°N - 30°S, and 30°S - 60°S. These particular zones were chosen to encompass a sufficient number of data points as to give some statistical validity to the deposition concentration - MAR relationship. In addition, it was assumed that in the 60°N - 90°N and 60°S - 90°S zones the strontium-90 deposition concentrations were the same as in the 30°N - 60°N and 30°S - 60°S latitude bands respectively. The average strontium-90 deposition concentrations were estimated for each zone and the world-wide burden of strontium-90 was calculated by substitution of the appropriate results in equation (1). The resulting calculated global deposit of strontium-90 on July 1, 1959 was 4.3 megacuries.

The essential difference between Method 3 and Method 1 (total burden of 3.6 megacuries of strontium-90) lies in the determination of the mean zonal deposition concentration. In Method 1 it is assumed that the strontium-90 deposition concentration is independent of rainfall and the unweighted average of the deposition concentrations is assumed to be the mean deposition concentration for the whole latitude band. In Method 3 the strontium-90 deposition concentration - MAR relationship is assumed to hold for the latitude band and the mean zonal deposition concentration of strontium-90 is found from the value corresponding to the MAR given by Möller for the latitude band.

It is believed that the assumption of a zonal deposition concentration-MAR relationship has more validity than the assumption that the strontium-90 deposition concentration is independent of the MAR within the latitude band. If, indeed, the deposition of strontium-90 is caused by rainfall (as it is believed to be for the most part), then Method 3 would probably be more valid for it takes this causality into account. An idea of the representativeness of the soil sampling can be gained by referring to Figure 7.2 in which the mean of the MAR from these samples is 43 inches for the 30°N - 60°N zone. Möller's value of 27 inches should be much closer to the actual mean for the zone since it is based on much more extensive sampling of precipitation throughout the band.

Alexander et al[1] - Method 4

A fourth procedure, adopted by Alexander and his coworkers, is similar to that used by Machta and List[7]. In principle this method is also similar to the analysis described under Method 3. Calculations of strontium-90 deposits, based on the results available from soil sampling sites, were superimposed on a climatological map of the world. The precipitation values, for areas where no soil sampling was conducted, were then used, in conjunction with

plots of strontium-90 deposition concentration as a function of precipitation to determine the activity per square mile. Integration of these strontium-90 deposits yielded the global deposition of this nuclide. A result of 4.1 megacuries of strontium-90 was calculated as being present on the surface of the earth on July 1, 1959. Calculations of the deposit by the same method for other years yielded 2.3 megacuries of strontium-90 for July 1, 1958 and 0.8 megacuries for July 1, 1956.

Alexander[8]--Method 5

The final method described here was actually the first and simplest method used by Alexander to determine the quantity of global fallout. It entails calculation of the average deposit within a given area from the simple arithmetic average of the strontium-90 measurements at the sampling sites within the area. The resultant average deposition, expressed in activity per square mile, is then multiplied by the area of the zone. The summation of the zonal deposits yields the total burden of strontium-90. Obviously rainfall data are not considered in this method and it is assumed that the sampling sites are randomly distributed within the given area with respect to precipitation type and amount. If this is not so, and the sites are situated in regions of heavy precipitation compared to the zonal average, the resultant integrated deposit of strontium-90 within this area will be high. Conversely the integrated deposit will be low if the sites are situated in relatively low rainfall regions.

This calculation was applied to the results of strontium-90 analyses of soils in 1959. A resultant global deposit of strontium-90 on July 1, 1959 of 6.2 megacuries was calculated.

Uncertainties in Sampling and Analysis

In this report we are concerned only with soil sampling sites since

very few ocean water measurements have been made. The first criterion for the choice of a suitable site is that all the radioactivity which has been deposited there has remained in the soil and that redistribution has been minimal. Representativeness of the samples used for analysis and possible errors in the analytical procedures must be considered in these studies. Many possible errors in the sampling and the chemical analysis of soil were discussed thoroughly by Alexander[1,8], and by others. It was concluded from these studies that the error in precision of obtaining a representative sample by methods employed by Alexander was less than 10%. However, as a result of the incomplete chemical extraction of strontium-90 from soils, the global integrals of strontium-90 are probably low by about 10%.

Comparison of the Global Integrals

The five methods used to calculate the global deposit of strontium-90 on July 1, 1959 gave these results:

Method 1	: 3.6 MC	Method 3	: 4.3 MC	Method 5	: 6.2 MC
Method 2	: 3.9 MC	Method 4	: 4.1 MC		

The high value yielded by Method 5 apparently resulted from a calculated value of 2.2 megacuries for the 20°N-30°N latitude band, wherein the majority of the sites were located in areas of extremely high precipitation. If a more realistic value is assigned to this region, fairly good agreement among all methods results.

Some of the small differences between the results of the different approaches can be explained. Methods 1 and 2 yielded lower deposits of strontium-90 than Methods 3 and 4, primarily because of the reasons outlined in the procedure for Method 3. The precipitation sites and the soil sampling sites are, on the

whole, located in areas which have higher precipitation amounts, on the average, than the estimates of the latitude zone average given by Möller. As a result of the inverse relationship between deposition concentration of strontium-90 and the amount of precipitation, the values obtained by Methods 1 and 2 will be lower than when this relationship is taken into account, as in the other two approaches. It is also remarkable that Methods 3 and 4 agree so well, since Method 3 is, indeed, a much simpler procedure than Method 4.

The question of what is the overall uncertainty in these calculations of the surface burden is an extremely difficult problem in itself. Added to the 10% underestimation of the strontium-90 inventory as a result of the chemical processes there are several other sources of uncertainty. There is, of course, the statistical question of the validity of the assumption that the results of so few analyses can be considered typical of the distribution of strontium-90 on the earth's surface. Not only is the number of analyses small, but in addition the sampling sites are restricted to the land with very little information being available on the amounts of strontium-90 in the oceans, which comprise 70% of the total surface of the earth. In all of the above methods it was tacitly assumed that strontium-90 was deposited in the oceans in much the same manner as on the land and, hence, the concentrations in the oceans, when integrated, would yield results similar to those found on the land for comparable areas. Unfortunately this assumption may be in serious error. The work of Bowen and Sugihara[9] has indicated that considerably more radioactivity may have been added to the oceans than what has hitherto been assumed in the above calculations. However, the results of this work are difficult to interpret until much more data are available.

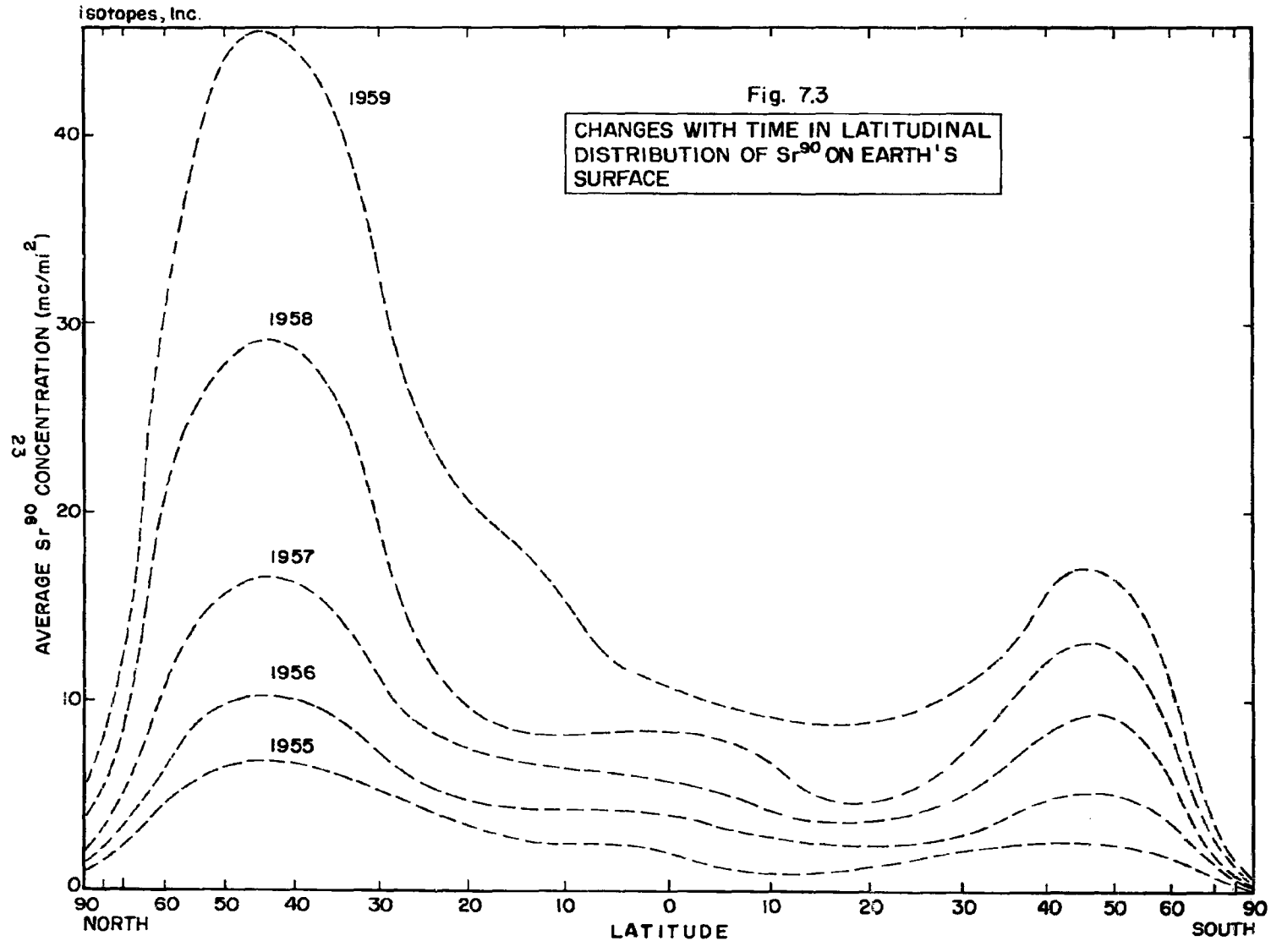
Also a point to be considered is the values chosen for the precipitation distribution patterns over the globe. Long term climatological means were

considered by Möller when he calculated average precipitation values for latitude bands and these results will, of course, vary from year to year. A study of this question has been made and it was concluded that the uncertainty from this source alone in the estimate of global fallout was between 20 and 30%[10].

It is clear, therefore, that it is difficult to devise a statistically sound basis for calculating the overall error in the estimates of the surface burden of strontium-90. Alexander et al[1], on the basis of several "reasonable" analyses, concluded that the "random uncertainty" in their 1959 integral of 4.1 megacuries of strontium-90 is about 40%. It appears that this is about the best estimate of the probable uncertainty which can be made at the present time without entering into an almost impossible and certainly a prohibitively expensive program of experimental investigation.

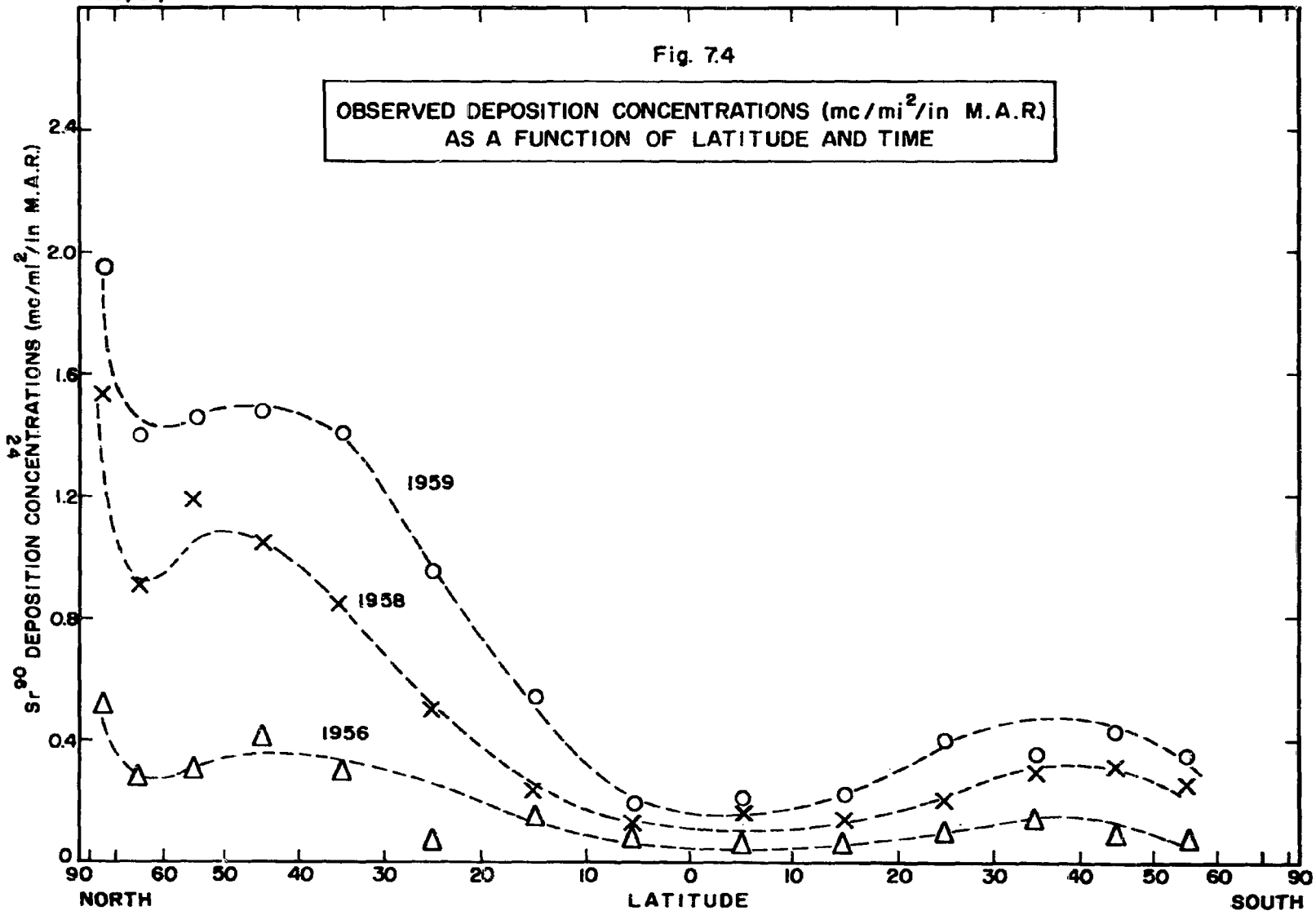
Distribution of Surface Fallout of Strontium-90

The general features of the distribution of strontium-90 on the surface of the earth, as shown in Figures 7.3 and 7.4 for the years from 1955 to 1959, have been well-known for some time and can be explained reasonably well by current theories of fallout mechanisms. The two major peaks in the strontium-90 concentration, which occur in the 30° - 50° latitude bands of both hemispheres, can be clearly seen; the highest value for a latitude zone (40°N - 50°N) in the Northern Hemisphere on July 1, 1959 was 45 mc/mi^2 and the highest value for a latitude zone (40°S - 50°S) in the Southern Hemisphere at the same time was about 17 mc/mi^2. These values are average surface concentrations for the latitude zones and the spread in concentrations for individual sites within a 10° latitude band is certainly a factor of five. Surface concentrations in 1959 reached as high as 100 mc/mi^2 at some sites in the 30°N - 50°N band, even in areas not greatly influenced by intermediate fallout from the Nevada tests, and in other areas the concentrations were as low as 10 to 20 mc/mi^2. This spread is


Isotopes, Inc.
Fig. 7.3
CHANGES WITH TIME IN LATITUDINAL DISTRIBUTION OF Sr^{90} ON EARTH'S SURFACE
AVERAGE Sr^{90} CONCENTRATION (mc/mi^2) $\times 23$
1959
1958
1957
1956
1955
0
10
20
30
40
90 60 50 40 30 20 10 0 10 20 30 40 50 60 90
NORTH
LATITUDE
SOUTH

Isotopes, Inc.

Fig. 7.4



largely governed by the range in annual precipitation from site to site. The latitudinal distribution of deposition concentrations as shown in Figure 7.4 provides the explanation for the variation of the surface concentration (mc/mi^2) of strontium-90 with latitude. At first sight it could be concluded that the distribution of strontium-90 with latitude as shown in Figure 7.3 was a result of the distribution of precipitation over the earth. However, Table 7.5, which gives the north-south distribution of rainfall, demonstrates that, in the main, precipitation amounts and fallout deposition are not related. Rather the north-south profile of the strontium-90 deposit is correlated with the north-south variability of strontium-90 concentrations in rainfall. This distribution of the concentrations of strontium-90 in rainfall is largely a result of meteorological circulations which transport the radioactivity from the stratosphere to the troposphere in preferred areas. These mechanisms are discussed in greater detail in Chapter 6.

It must be borne in mind that Figures 7.3 and 7.4 were derived from the analysis of soil and, hence, they represent what is believed to be the distribution of strontium-90 over the land masses. Some of the details of these curves, such as the trough in the curve in Figure 7.3 just south of the equator, and some of the apparent minor changes in the shapes of the curves from year to year are not given much weight. A discussion of these trends is possible only when a dense network of observation stations, akin to that in the U. S., is in operation. Moreover, these curves are likely to be much smoother if the deposition of fallout in the oceans is taken into account. What data are available at the present time on strontium-90 in the oceans[9] do tend, however, to support the overall distribution picture.

Deposition of Strontium-90 in Polar Regions

There has been a certain amount of speculation on the deposit of

strontium-90 in polar regions because of the shortage of good samples from these areas. Data on radioactivity in snow and ice-cores collected prior to 1958 suggest that the concentration of strontium-90 in precipitation in these regions was not greatly different from that observed in adjacent latitude zones. Further supporting evidence can be found in the radioactivity measurements in ground-level air filter samples, which suggest once again only small differences in the total beta activity between the results from the 50°N to 60°N latitude region and the data from latitudes higher than 60°[5]. It is likely, however, that during the latter part of 1958, when the U. S. S. R. conducted several high yield nuclear tests in the Arctic, that concentrations of strontium-90 in both air and precipitation in this region would be higher than in the 50°N - 60°N latitude band. In Methods 1, 2 and 3 for the calculation of global fallout it was assumed that the concentrations of strontium-90 in precipitation and the deposition concentrations in soils in the 30° - 90° latitude bands of both hemispheres were constant. Because of the data on air and precipitation samples discussed above, it is believed that this assumption does not introduce a serious uncertainty in the total surface burden of strontium-90. Moreover, to change the ground-level burden of strontium-90 by just 10% would necessitate employing concentrations, for the most northerly and southerly latitudes, at least an order of magnitude higher than those used. This is believed to be extremely unlikely even after allowing for the contribution from Soviet testing in late 1958.

Finally, it is understood that precipitation amounts are not accurately known for Arctic and Antarctic areas. However, global energy considerations and other data suggest that the values of precipitation assigned to the polar regions are not uncertain by amounts which could seriously alter present estimates of the inventory of strontium-90 on the earth's surface.

Incremental Deposits of Strontium-90

The annual increments of strontium-90 added to the earth's surface since the commencement of testing are worthy of note (see Table 7.7). During 1955-56 and 1956-57 the total global increment of strontium-90 was between 0.4 and 0.5 megacuries in each year. These activity increments rose to 0.9 megacuries in 1957-58 and to 1.3 megacuries in 1958-59, and then dropped to about 0.6 megacurie in 1959-1960, reflecting the increased number of nuclear tests performed during 1957 and 1958 and the moratorium which commenced in late 1958. During the period 1955 to 1959 the fraction of the total deposit which was delivered to the Northern Hemisphere rose quite rapidly (Table 7.8). Thus during the period 1955-1956 only about 35% more strontium-90 was deposited in the Northern Hemisphere than in the Southern Hemisphere, compared to approximately three times as much during the period 1958-1959. As discussed by Walton[6], the data on fission products in precipitation during the latter period indicate a high contribution of strontium-90 from October 1958 Soviet tests in the deposit. These cursory examinations of strontium-90 in soils confirm that nuclear debris from Soviet tests is largely confined to northerly latitudes whereas debris from U. S. and U. K. shots at near equatorial latitudes is more evenly distributed over the globe.

Material Balance Studies

It has been estimated that 5.0 to 5.5 megacuries of strontium-90 had been injected into the stratosphere by the end of the autumn 1958 Soviet tests (see Chapter 5). Calculations of the surface burden of strontium-90 by the methods described in previous sections yielded values for July 1, 1959 of between 3.6

Table 7.7. Calculated Annual Increments to the Surface Burden of Strontium-90 (Megacuries)

Period	Method 1 Data From Soil Analyses	Method 2 Data From Precipitation Analyses
Prior to 1 July 1955	0.52	0.72
1 July 1955 - 1 July 1956	0.40	0.63
1 July 1956 - 1 July 1957	0.47	0.54
1 July 1957 - 1 July 1958	0.92	0.63
1 July 1958 - 1 July 1959	1.32	1.37
1 July 1959 - 1 July 1960	---	0.58

Table 7.8. Variation with Time of the Strontium-90 Distribution Between the Hemispheres

Time	Megacuries of Sr^{90} (Method 1)		Ratio of Annual Increments
	N. Hemisphere	S. Hemisphere	
July 1, 1955	0.38	0.14	
July 1, 1956	0.61	0.31	1.35
July 1, 1957	0.91	0.48	1.76
July 1, 1958	1.58	0.73	2.58
July 1, 1959	2.58	1.05	3.12
July 1, 1960 *	2.95	1.26	1.75

* Rainfall increments added to soil data for 1959.

and 4.3 megacuries (excluding Method 5). Probably a reasonable average of these data is 4.0 megacuries. It was also estimated in Chapter 6 that the stratospheric burden on July 1, 1959 was about 1.1 megacuries of strontium-90. Some radioactive decay of the total of 5.0 to 5.5 megacuries mentioned above would have occurred, but this is estimated to be less than 10%.

Obviously in spite of our reservations concerning the accuracy of the ground-level burden, there is excellent agreement between the "observed" sum of the surface burden of strontium-90 plus the stratospheric inventory and the estimated total stratospheric injection. It might be pointed out that it has been tacitly assumed that the ground-level burden of strontium-90 as calculated for July 1, 1959 was purely stratospheric in origin. This is not unreasonable since the results from those sites which were affected by the Nevada testing have been omitted from the calculation and the contribution of world-wide tropospheric fallout from high yield detonations is certainly less than 10% of the stratospheric injection.

Future Deposition Levels of Strontium-90

It is understood that any prediction of the future burdens of strontium-90 on the earth's surface will be more uncertain than estimates of the present deposit because of errors which will arise in the choice of models of stratosphere-troposphere interchange used to describe the rate of deposition of nuclear debris which remains in these reservoirs. A one-block model of the stratosphere, with first-order kinetics being applied to the rate of release of strontium-90 from the stratosphere to the troposphere is the most popular model used to date. In the present study a different approach was used. Four sections of the stratosphere

were assumed, the tropical stratosphere (a) up to 70,000 feet, where HASP measurements were plenteous, (b) between 70,000 and 90,000 feet, where balloon flight data are the only sources of information, and (c) above 90,000 feet, where no direct measurements were made, and (d) the polar stratosphere. All sections are considered separately and it is assumed that first-order kinetics can be applied to each. For the tropical lower altitude section the residence half-time, $t_{1/2}$ = 1 year, for the second section $t_{1/2}$ = 3 years, and for the highest altitude portion $t_{1/2}$ = 5 years. For the polar section, $t_{1/2}$ = 3 months. The following mathematical analyses were then applied to the five areas under consideration, i.e., to the material already deposited on the earth's surface by July 1, 1959, and to the material still in each of the stratospheric sections on that date.

The deposit representing strontium-90 already on the earth's surface on July 1, 1959 is given by

$$S_s = S_o e^{-\lambda t} .$$

where S_o = strontium-90 activity on the surface on July 1, 1959,
S_s = strontium-90 activity on the surface at a time, t, later,
λ = decay constant of strontium-90.

The deposit of strontium-90 on the ground at a time, t, after 1959 as a result of the fallout of debris residing in the lower, intermediate, or high stratosphere on July 1, 1959 requires a somewhat more complicated expression. Thus two processes lead to the decrease of the strontium-90 concentrations in the stratosphere - radioactive decay and fallout to the troposphere and to the ground. The quantity of strontium-90, P, remaining in the polar stratosphere,

the quantity, L, remaining in the lower tropical stratosphere (below 70,000 feet), the quantity, I, remaining in the intermediate layer, and the quantity, U, remaining in the upper stratosphere may be expressed by

$$P = P_o \exp(-\lambda_p t)\exp(-\lambda t),$$
$$L = L_o \exp(-\lambda_l t)\exp(-\lambda t),$$
$$I = I_o \exp(-\lambda_i t)\exp(-\lambda t),$$
$$U = U_o \exp(-\lambda_u t)\exp(-\lambda t),$$

where P_o, L_o, I_o and U_o were the quantities of strontium-90 in the various regions of the stratosphere on July 1, 1959, and λ_p, λ_l, λ_i and λ_u are the removal rate constants for each of these regions, and are equal to 0.693 divided by $t_{1/2}$ for the region. By assuming that all material which has fallen out of each region of the stratosphere by time, t, has reached the surface by that time, we obtain the following expressions for the quantities of strontium-90, P_s, L_s, I_s and U_s, from each region of the stratosphere which are on the surface at time t:

$$P_s = P_o \left[1-\exp(-\lambda_p t)\right] \exp(-\lambda t) \quad (3)$$
$$L_s = L_o \left[1-\exp(-\lambda_l t)\right] \exp(-\lambda t) \quad (4)$$
$$I_s = I_o \left[1-\exp(-\lambda_i t)\right] \exp(-\lambda t) \quad (5)$$
$$U_s = U_o \left[1-\exp(-\lambda_u t)\right] \exp(-\lambda t) \quad (6)$$

The total surface burden, S, during years following 1959 is obtained by summing equations (2), (3), (4), (5) and (6):

$$S = S_s+P_s+L_s+I_s+U_s$$
$$S = \left[S_o + P_o(1-e^{-\lambda_p t})+L_o(1-e^{-\lambda_l t})+I_o(1-e^{-\lambda_i t})+U_o(1-e^{-\lambda_u t})\right]e^{-\lambda t} \quad (7)$$

In Chapter 6 certain values of λ_p, λ_l, λ_i and λ_u were adopted to obtain correspondence between estimates of stratospheric injections and HASP calculations of stratospheric burdens. As a result, the values obtained for the various constants in equation (7) are:

S_o = 4.0 megacuries of strontium-90
P_o = 0.18 " " "
L_o = 0.30 " " "
I_o = 0.50 " " "
U_o = 0.15 " " "
λ = 0.247 yr^{-1}
λ_p = 3.08 yr^{-1}
λ_l = 0.693 yr^{-1}
λ_i = 0.231 yr^{-1}
λ_u = 0.139 yr^{-1}

The results of this calculation are shown in Table 7.9 and in Figure 7.5. Two general features are clearly observable in the curve of the predicted total ground-level burden of strontium-90. Firstly, it appears that the peak deposition will occur in 1961-1962, and secondly, this peak will be followed by a gradual decrease in the total burden. This decrease should closely follow the radioactive decay rate of strontium-90 after about 1970. As discussed in a previous section the absolute value of the total deposit of strontium-90 is in some doubt, but the general features of the curve in Figure 7.5 should be nearly independent of this fact. The values obtained here for future strontium-90 burdens may be used in assessing the dosage to be expected from strontium-90 during the years to come, provided weapon testing in the atmosphere is not resumed.

The foregoing calculation has neglected the effect of debris from the rocket shots, Teak and Orange. It is not at all likely that fallout from these weapons will materially change the projected surface burdens, however. A reasonable estimate of its effect is that the peak in the burden will occur in 1962-1963, about one year later than calculated above, and that the burden will be slightly higher, about 0.07 megacurie higher, than was calculated.

Table 7.9. Future Surface Burdens of Strontium-90 from Worldwide Fallout

Date	Calculated Total Deposit (Megacuries)	
	If Weapon Testing is not Resumed	If Weapon Testing is Resumed*
1 July 1960	4.2	4.2
1 July 1961	4.27	4.27
1 July 1962	4.27	5.03
1 July 1963	4.24	6.10
1 July 1964	4.19	7.24
1 July 1965	4.12	8.38
1 July 1966	4.05	9.53
1 July 1967	3.97	10.67
1 July 1968	3.89	11.79
1 July 1969	3.81	12.90
1 July 1970	3.72	13.99

* Assuming an injection of 1.0 MC into the polar stratosphere on the first of January of each year, beginning 1 January 1962, and an injection of 0.5 MC into the tropical stratosphere on the first of July of each year, beginning 1 July 1962.

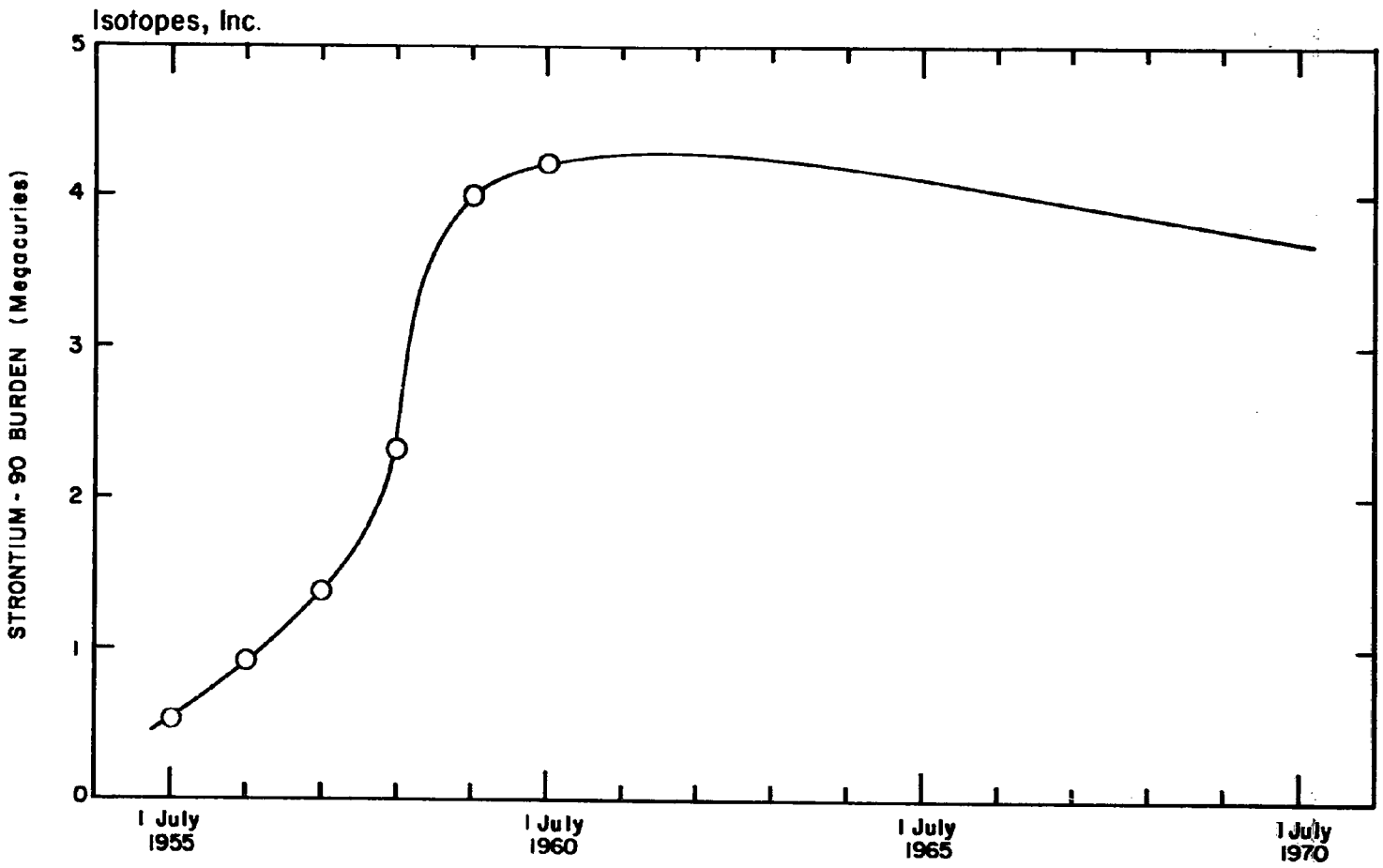


FIG. 7.5 MEASURED PAST BURDENS AND PREDICTED FUTURE BURDENS OF STRONTIUM-90 ON THE SURFACE OF THE EARTH

Should weapon testing in the atmosphere be resumed, the surface burden of strontium-90 would again begin to rise. The rate of increase of the burden would depend upon the rate of weapon testing, of course, and it cannot be predicted with any certainty. If the rate of testing approached that which prevailed during 1958, the surface burden could reach quite a high value.

As an example of one possible situation, we have assumed that testing would be resumed by the beginning of 1962, and that one megacurie of strontium-90 would be injected into the polar stratosphere on about the first of January each year, beginning on 1 January 1962, and that 0.5 megacuries would be injected into the tropical stratosphere on about the first of July each year, beginning on 1 July 1962. Using the same method of calculation of future burdens which gave us the data in the second column of Table 7.9, but with these new assumptions, we obtain the estimates in the third column of that table. Under these conditions the stratospheric burden would stabilize in an equilibrium range between 2.1 and 3.2 megacuries, varying from season to season in response to fallout and to new injections. By 1993 the fallout rate would approach a value of about 1.44 megacuries per year reaching the ground. The surface burden would ultimately approach a value of about 60 megacuries if this fallout rate were maintained indefinitely. It would reach 95 percent of this value by about 2083, one hundred and twenty one years after the assumed resumption of testing.

Summary

Five methods for the determination of the total amount of strontium-90 on the earth's surface are presented. Good agreement among these different approaches is observed. Four of the methods yield global integrals of strontium-90 for July 1, 1959 ranging from 3.6 to 4.3 megacuries. It is concluded that the average of 4.0 megacuries of strontium-90 is probably the best estimate of the surface burden for mid-1959. By July 1960 the burden had increased by an additional 0.6 megacuries.

These estimates of the surface burden have associated with them an error of at least $\pm$ 40 percent. Two major uncertainties contribute to this error: the amount and distribution of precipitation over the globe and the quantities of fallout in the oceans.

Fallout strontium-90 is distributed unevenly between and within the hemispheres. By July 1, 1959, 2.5 times as much strontium-90 had been deposited in the Northern Hemisphere as in the Southern Hemisphere, thus reflecting the exclusive deposition of Soviet debris in the former. Excessive deposits of strontium-90 occur in the middle latitudes of both hemispheres as a result of the meteorological factors governing the release of nuclear debris from the stratosphere.

Material balance studies also exhibit remarkably good agreement between estimated stratospheric injections of strontium-90 and the amounts of this nuclide present on the earth's surface at various times.

Predictions of future ground-level burdens indicate that, unless weapon testing is resumed, a peak deposit of strontium-90 will be attained in 1961-1962 followed by a slow decrease in the burden according to the half life of strontium-90.

INTERMEDIATE-RANGE FALLOUT FROM NEVADA TESTS

At the present time it is convenient to divide fallout into three types:

(a) local fallout--rapidly deposited within a few miles of the test site.

(b) intermediate fallout--fairly rapidly deposited over an area hundreds of miles from the test site, and

(c) long-range fallout--which may be further subdivided into tropospheric and stratospheric depending upon whether the deposited material actually entered the stratosphere or not. This type requires time periods ranging from days to several years for its deposition on a world-wide basis.

Since 1954 considerable effort has been devoted to determining the extent and magnitude of both local and long-range fallout, but relatively little work has been directly performed in examining intermediate fallout. It is now possible, however, from observations of strontium-90 concentrations in soils both in the U. S. and at other stations outside the U. S. but in the same latitude band as the Nevada test site, to estimate the amount of intermediate fallout of strontium-90 around the test site. The area encompassed by this type of fallout also becomes clearly apparent from this examination.

Some of the possible applications of this kind of study include the assessment of fallout and the hazard resulting from a nuclear war. It also serves to define which soil samples can be included when the surface burden of strontium-90 of stratospheric origin is estimated. This is of immediate application to the study described in the previous section of this report.

This analysis has been described also in a recent report on studies of nuclear debris in precipitation[2].

Procedure

The procedure used in this investigation depends upon the fact that the strontium-90 deposition concentrations in soils (mc/mi^2/inch MAR)

within the intermediate fallout zone will be higher than at comparable sites outside the zone. A simple investigation of the total deposit (mc/mi^2) will not suffice to define this zone because it is difficult to distinguish areas which have high strontium-90 concentrations because they received local and intermediate fallout from Nevada from other areas which have high concentrations of strontium-90 because they are situated in regions of high rainfall. One complicating factor does arise, however, when only the deposition concentrations are considered. This factor is clear from the inverse correlation which exists between the strontium-90 deposition concentrations for soils and the total precipitation amounts.

It is observed that the correlation between these two variables results in areas with low precipitation possessing higher strontium-90 deposition concentrations than regions experiencing larger precipitation amounts. This relationship is clearly visible from the data which have been plotted in Figure 7.1 for the soil results obtained by Alexander et al in 1958. Using this curve, together with known precipitation amounts at several sites throughout the U. S. it is then possible, however, to predict what the soil concentration should be if it had not been affected by intermediate fallout. Subtraction of the predicted amount from the observed amount, followed by multiplication of this difference by the precipitation observed at the site since fallout commenced (say 1953) yields the excess due to fallout from sources other than the stratosphere. This remainder is primarily intermediate fallout,and the total contribution from this source within the U. S. may then be determined by integration of the excess amounts.

The results of this calculation for the measurements performed in 1958 are shown in Figure 7.6 where the excess quantities of strontium-90 are indicated on a map of the U. S. Isopleths of these excess deposits of

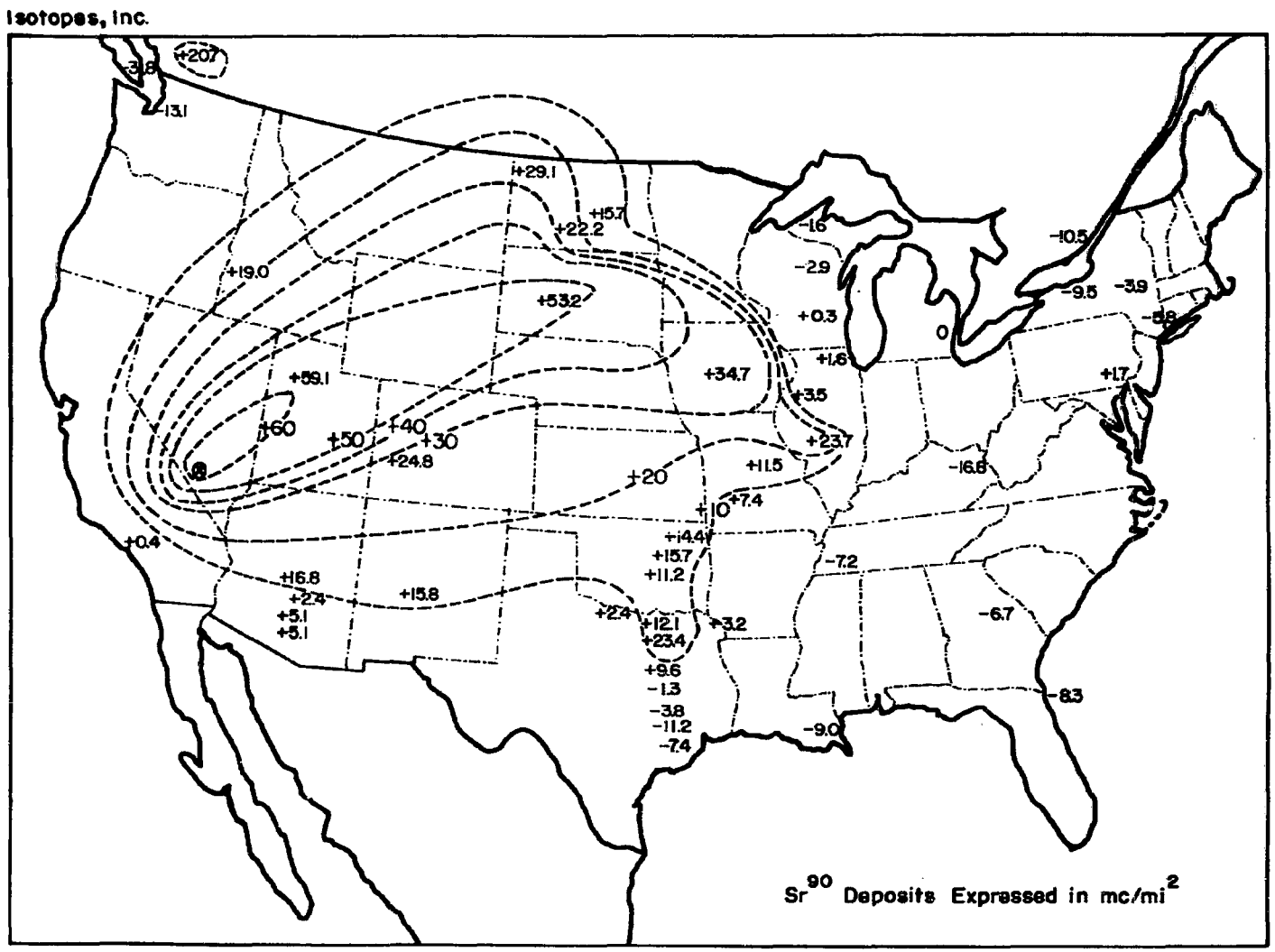


FIG. 7.6 STRONTIUM-90 IN U.S. SOILS FROM INTERMEDIATE FALLOUT

strontium-90 were drawn from the above points.

To determine the total amount of intermediate fallout within the U. S., it was decided to integrate above the + 10 mc/mi^2 isopleth to allow for any small deviations from the expected average conditions. A total excess deposit of 40.9 kilocuries of strontium-90 attributable to intermediate fallout was calculated.

Discussion

The results calculated above may be compared to data available on the magnitude of the testing carried out in Nevada and an independent estimate of the same quantity recently reported by List[11]. According to released reports, the fission yield of weapons tested in Nevada up to July 1, 1958 was about 970 kilotons[11], which is considered to be equivalent to the production of close to 100 kilocuries of strontium-90. Thus approximately 40% of this material can be accounted for by intermediate fallout. The majority of the remainder, presumably, came down as local fallout. List[11], in his calculation, considered only the actual deposit (in mc/mi^2) in the U. S. and to determine the excess from Nevada tests he subtracted the contribution from the stratospheric source which he obtained by estimation of the average deposition within the same latitude band outside the U. S. An estimate of the burden from stratospheric sources was 40 mc/mi^2 and the total activity of strontium-90 above this "isoline" was calculated by List for the U. S. to be 23.2 kilocuries. List also calculated that the excess strontium-90 above the 30 mc/mi^2 isopleth was 47.8 kilocuries.

Considerable uncertainties are inherent in the above methods of calculation and perhaps one of the greatest lies in the subjective manner in which certain isopleths of excess concentration are drawn. On the basis of gummed film data we might have expected about 30 kilocuries of strontium-90

or slightly less for intermediate fallout compared with List's estimate of 23.2 kilocuries and our determination of 41 kilocuries. Certainly the estimate given in this chapter has an uncertainty of at least 25% and we might conclude, therefore, that an average of 30 kilocuries of strontium-90 is a fairly good measure of intermediate fallout in the U. S. Without many more soil stations and strontium-90 data in the United States, it is unlikely that a much better determination can be made with present information.

FALLOUT OF TUNGSTEN-185

The assignment of nuclear debris sampled at the earth's surface to a particular explosion of a nuclear device is extremely valuable in any attempt to determine the mechanism of fallout. Unfortunately this was rather difficult with most of the tests conducted up to mid-1958 even though analyses of short-lived fission product nuclides were of some assistance in this respect. During the Hardtack test series of the spring and summer of 1958, however, tungsten-185, a nuclide which is not normally produced in any great quantity during fission or fusion, was produced by some of the devices. This nuclide has a half life of 74 days. Thus a unique opportunity was presented which permitted debris to be assigned to devices which were detonated at a known time and in a known area. Tungsten-185 was subsequently determined both in stratospheric air samples and in precipitation samples collected at ground-level. A review of the data reveals that insufficient advantage was taken of this valuable tool. The data for precipitation samples are very scanty and insufficient coverage both in time and area is apparent from the results[12]. It is possible, however, to follow the deposition on the earth's surface from September 1958 through December 1959 and to estimate the

overall burden for this period. Sampling was even more meager for the period prior to September 1958, when tropospheric fallout of tungsten-185 probably predominated, and only a crude approximation can be made of the deposition at this time. Even during the more intense sampling period which followed, it is unlikely that the total deposition amounts assigned to this nuclide are accurate to within a factor of two. Furthermore, we are confronted with grave uncertainties in the total yield of tungsten-185 from the shots as well as in the fraction of the yield which was injected into the stratosphere. Libby[13] estimated a total yield of tungsten-185 of 250 megacuries, corrected for decay to August 1, 1958. Martell and Drevinsky[14] assumed that 40% of this amount entered the stratosphere. On the other hand we assumed (Chapter 6) that 95 megacuries of tungsten-185 were injected into the stratosphere. Corrected for decay and for fallout, the burden was 53 megacuries on August 15, 1958. If all the above estimates are adjusted to a reference date of June 1, 1958, our estimate becomes 108 megacuries of tungsten-185, which may be directly compared with the Libby-Martell estimate of 177 megacuries. This date is chosen at this stage of the discussion because all precipitation data from the AEC world-wide network of sampling stations were adjusted to this time. There is obviously a considerable difference between these estimates, thus adding further uncertainties to evaluations of the rate of fallout of this nuclide. Very few data are available on the amount of local fallout of tungsten-185 which may possibly be used to check the stratospheric injection amounts.

Method of Calculation

The procedure employed for the calculation of the surface burden of tungsten-185 is similar to Method 2, which describes the computation of global fallout of strontium-90. Average quarterly concentrations of tungsten-185 in

precipitation in 10° latitude bands were calculated, and these data were multiplied by the precipitation for the particular band during the three-month period and the area of the band to determine the total deposit for the quarter. Precipitation values adopted for the three-month period were one-quarter of the mean annual rainfalls which were given by Möller[4]. Some interpolation of the results was necessary because of lack of data and in some cases because of anomalous results. These are indicated in the tables which follow.

Results

Table 7. 10 lists calculations of quarterly concentrations of tungsten-185 in precipitation at each of the rain water stations operated by the U. S. AEC[12]. These concentrations were calculated from the monthly values obtained at each station by dividing the total deposit for the three-month period by the total precipitation for the same time period. Complete records are available for only a few of these sites and, in cases where only one or two month's record is at our disposal for any one quarter, this value was assumed to be applicable to the whole of the three-month period. For the third quarter of 1958 results are available for most stations for the month of September only. The total deposit was calculated for just one month for this time interval. Relatively few stations were monitoring tungsten-185 during 1958. Many more, particularly in the United States, were in operation during 1959, but the world-wide coverage was still poor. Only two stations were monitoring the Southern Hemisphere beyond 40°S, and in the Northern Hemisphere there were only four beyond 50°N. From the results shown in Table 7. 10 the average concentrations in each 10° latitude band were determined and are shown in Table 7. 11. Certain stations (those indicated by an asterisk in Table 7. 10) were omitted from these calculations. In the examples which were rejected, extremely high tungsten-185 concentrations, orders of magnitude higher than

Table 7.10. Average Quarterly Concentration of Tungsten-185 in Precipitation (mc/mi/inch)**

Station	Latitude (degrees)	1958		1959			
		3	4	1	2	3	4
70° - 80° N							
Barrow, Alaska	71	---	---	---	520	16.8	11.2
Thule AFB, Greenland	77	---	---	---	---	---	34.0
60° - 70°N							
Oslo, Norway	60.5	---	---	---	---	20.6	2.6
Anchorage, Alaska	61	---	---	---	60.0	13.6	19.1
50° - 60°N							
Prestwick, Scotland	57	---	---	---	---	---	7.5
Cold Bay, Alaska	55	---	---	3.7	41.2	23.8	2.4
Juneau, Alaska	58	---	---	---	68.0	5.1	0.6
40° - 50°N							
Williston, N. D.	49	---	---	---	280.0	---	7.9
International Falls, Minn.	48	---	---	---	63.8	3.9	6.2
Seattle, Wash.	48	160.5*	8.7	26.8	77.9	6.0	3.7
Vienna, Austria	48	16.7	18.2	39.0	48.5	3.8	---
Helena, Mont.	47	---	---	23.8	89.6	100.0	9.6
Klagenfurt, Austria	47	15.2	10.9	---	119.4	26.3	5.2
Milan, Italy	45	---	---	---	---	---	2.2
Vermillion, S. D.	44	16.8	92.1	61.2	16.7	14.6	12.0
Green Bay, Wis.	44	---	---	---	31.8	8.8	5.4
Florence, Italy	43	---	---	---	---	16.0	---
Medford, Ore.	42	---	---	---	114.0	125.0	15.7
Salt Lake City, Utah	41	---	198	148.0	153.5	11.1	133.0
Lemont, Ill.	41	---	25.4	5.7	18.5	---	---
Pittsburgh, Penn.	41	12.8	27.1	35.6	60.3	12.9	28.8
New York City, N. Y.	41	---	---	---	75.3	9.6	1.7
Westwood, N. J.	41	---	---	43.7	48.8	8.3	7.5
30° - 40°N							
Columbia, Mo.	39	---	---	41.3	39.0	4.4	1.5
Denver, Colo.	40	---	---	21.4	125.0	31.9	69.3
Lajes Field, Azores	38	---	---	---	---	---	7.1
Louisville, Ky.	38	---	---	---	57.3	8.1	10.2
Richmond, Cal.	38	140*	10.1	---	---	---	---
San Francisco, Cal.	37	---	---	---	541.0*	---	---
Tulsa, Okla.	37	33.2	291	82.9	22.5	9.9	2.9
Hiroshima, Japan	36	19.9	0.8	25.6	13.0	12.8	2.4
Columbia, S. C.	34	---	---	---	17.6	2.3	1.3
Nagasaki, Japan	34	16.9	19.6	21.2	10.6	5.2	3.5
W. Los Angeles, Cal.	34	517*	46.6	37.9	808*	250*	---
Birmingham, Ala.	33	23.0	37.0	41.3	42.5	26.8	0.8
Dallas, Texas	33	---	---	27.1	29.8	14.3	6.0
El Paso, Texas	33	---	---	---	149.0	---	27.6
Beirut, Lebanon	32	---	---	---	---	---	7.6
Tachikawa, Japan	36	---	---	---	---	---	2.1
Sidi Slimane, Morocco	34	---	---	---	---	---	4.9
20° - 30°N							
Houston, Texas	29	21.2	28.7	30.6	31.2	10.9	6.6
Coral Gables, Fla.	27	37.8	13.5	32.8	13.8	1.6	---
Karachi, Pakistan	26	---	---	---	---	56.6	24.7
Taipei, Taiwan	24	7.2	22.1	51.8	10.4	1.1	2.3
Hilo, Hawaii	22	---	---	8.7	---	---	---
Oahu, Hawaii	22	48.2 11.9	26.2 2.8	42.3	25.2	4.3	1.1
Iwo Jima	25	---	---	---	---	3.8	5.6

Table 7.10 (continued)

Station	Latitude (degrees)	1958		1959			
		3	4	1	2	3	4
10° - 20°N							
Bangkok, Thailand	14	1.4	5.7	143.0	12.8	1.6	1.0
Wake Island	19	---	---	---	24.2	16.5	9.3
Mexico City, Mexico	19	---	---	---	---	46.1	0.8
Mauna Loa, Hawaii	19	---	---	6.1	125.0	12.7	6.4
0° - 10°N							
Canal Zone	9	---	---	---	---	4.6	1.0
Turrhalba, Costa Rica	10	---	---	---	---	2.2	---
Monrovia, Liberia	6	---	---	---	---	2.7	10.9
Lagos, Nigeria	6	---	---	---	---	0.9	4.3
0° - 10°S							
Nairobi, Kenya	2	---	---	---	---	---	3.1
Kikuyu, Kenya	2	---	8.4	6.4	32.6	20.8	8.8
Manaus, Brazil	3	---	---	4.0	1.6	5.1	---
Canton Island	3	---	---	2.5	27.2	10.1	49.1
Leopoldville, Congo	4	---	---	---	---	---	6.3
10° - 20°S							
Suva, Fiji	18	---	---	---	---	2.7	---
Lima, Peru	16	---	---	---	---	58.2	---
Darwin, Australia	17	---	---	2.1	---	---	---
Salisbury, S. Rhodesia	18	---	11.7	6.6	20.7	75.0*	1.4
Townsville, Australia	19	186	11.1	2.6	---	---	---
Guam	13	---	---	---	---	0.4	1.9
20° - 30°S							
Pretoria, Australia	25	---	2.2	22.6	13.1	---	25.3
Brisbane, Australia	27	---	---	2.6	---	---	---
Durban, Australia	29	3.2	37.2	9.3	26.1	1.8	6.8
30° - 40°S							
Perth, Australia	32	17.8	12.2	43.5	---	---	---
Santiago, Chile	33	9.7	---	15.5	12.5	10.1	97.3 *
Adelaide, Australia	35	---	---	86.2	---	---	---
Sydney, Australia	35	11.6	15.2	4.3	---	---	---
Melbourne, Australia	38	2.8	7.0	14.8	---	---	---
40° - 50°S							
Wellington, New Zealand	41	---	---	---	---	---	4.4
Hobart, Tasmania	42	---	---	21.6	---	---	---

* These data were omitted in calculating average values.

** All data adjusted for radioactive decay to June 1, 1958.

the remaining results, were observed probably because of very low rainfalls experienced at the stations. It was felt that the inclusion of these data would lead to an unduly high average concentration for the latitude band. In addition Table 7.11 indicates the number of results contributing to the average for the band. As with the strontium-90 data, average concentrations for the 30^{o} - 90^{o} bands have been determined from the available observations. These averages were obtained by weighting the mean tungsten-185 concentration in each 10^{o} latitude band according to the number of observations in that band. Table 7.12 lists the resulting idealized concentrations which were used to determine the ground-level deposit of tungsten-185 for the following times: October 1, 1958, January 1, 1959, April 1, 1959, July 1, 1959, October 1, 1959 and January 1, 1960. In Table 7.13 and Figure 7.7 the final calculations of the ground-level burden for these various times are shown. Summation of the world-wide deposits for each time interval (last line of Table 7.13) yields a total deposit of 155 megacuries by December 31, 1959. As pointed out previously, this result was calculated using a reference date of June 1, 1958. If adjusted to August 15, 1958, the integral becomes 77 megacuries of tungsten-185, which may be compared with the results in Chapter 6.

Discussion of Results

The change in ground-level burden of tungsten-185 with time, as shown in Figure 7.7 and Table 7.13, is interesting from several standpoints. It is clear first of all that a large fraction of the total amount of tungsten-185 was deposited during the spring of 1959, a result which is comparable with the data on strontium-90 deposition during the same period. The majority of the tungsten-185 was deposited at this time in the Northern Hemisphere during the annual "spring-rise" in fallout-deposition rate. Thus the spring-rise, as evident from fallout in the northern latitudes involves material injected into the equatorial stratosphere together with debris injected at higher latitudes. It must, therefore, be a meteorological phenomenon, and not entirely dependent

Table 7.11. Average Quarterly Concentration of Tungsten-185 in Precipitation in 10° Latitude Bands (mc W^{185}/mi^2/inch adjusted to June 1, 1958)

Latitude Band	1958		1959			
	Sept.	4	1	2	3	4
70°-80°N	---	---	---	520(1)	16.8(1)	22.7(2)
60°-70°N	---	---	---	60(1)	17.1(2)	10.8(2)
50°-60°N	---	---	3.7(1)	54.6(2)	14.4(2)	3.5(3)
40°-50°N	15.4(4)	54.3(7)	48.0(8)	85.6(14)	26.6(13)	18.4(13
30°-40°N	23.2(4)	56.7(6)	37.3(8)	50.6(10)	12.9(9)	10.5(14)
20°-30°N	24.0(4)	19.7(4)	33.2(6)	20.2(5)	13.0(6)	8.0(6)
10°-20°N	1.4(1)	5.7(1)	74.6(2)	54.0(3)	19.2(4)	4.4(4)
0°-10°N	---	---	---	---	2.6(4)	5.4(3)
0°-10°S	---	8.4(1)	4.3(3)	20.5(3)	12.0(3)	16.8(4)
10°-20°S	186 (1)	11.4(2)	3.8(3)	20.7(1)	20.4(3)	1.6(2)
20°-30°S	3.2(1)	19.7(2)	11.5(3)	19.6(2)	1.8(1)	16.0(2)
30°-40°S	10.5(4)	11.5(3)	32.9(5)	12.5(1)	10.1(1)	---
40°-50°S	---	---	21.6(1)	---	---	4.4(1)

Table 7.12. Idealized Tungsten-185 Concentrations* in Precipitation During 1958 and 1959

Latitude Band	1958		1959			
	Sept.	4	1	2	3	4
30°-90°N	19.3	58.2	40.4	85.5	20.1	13.8
20°-30°N	24.0	19.7	33.2	40.0**	13.0	8.0
10°-20°N	17.0**	5.7	20.0**	20.0**	19.2	4.4
0°-10°N	10.0**	7.0**	10.0**	18.5**	2.6	5.4
0°-10°S	10.0**	8.4	4.3	20.5	12.0	16.8
10°-20°S	10.0**	11.4	3.8	20.7	20.4	1.6
20°-30°S	3.2	19.7	11.5	19.6	1.8	16.0
30°-90°S	10.5	11.5	31.0	12.5	10.1	4.4

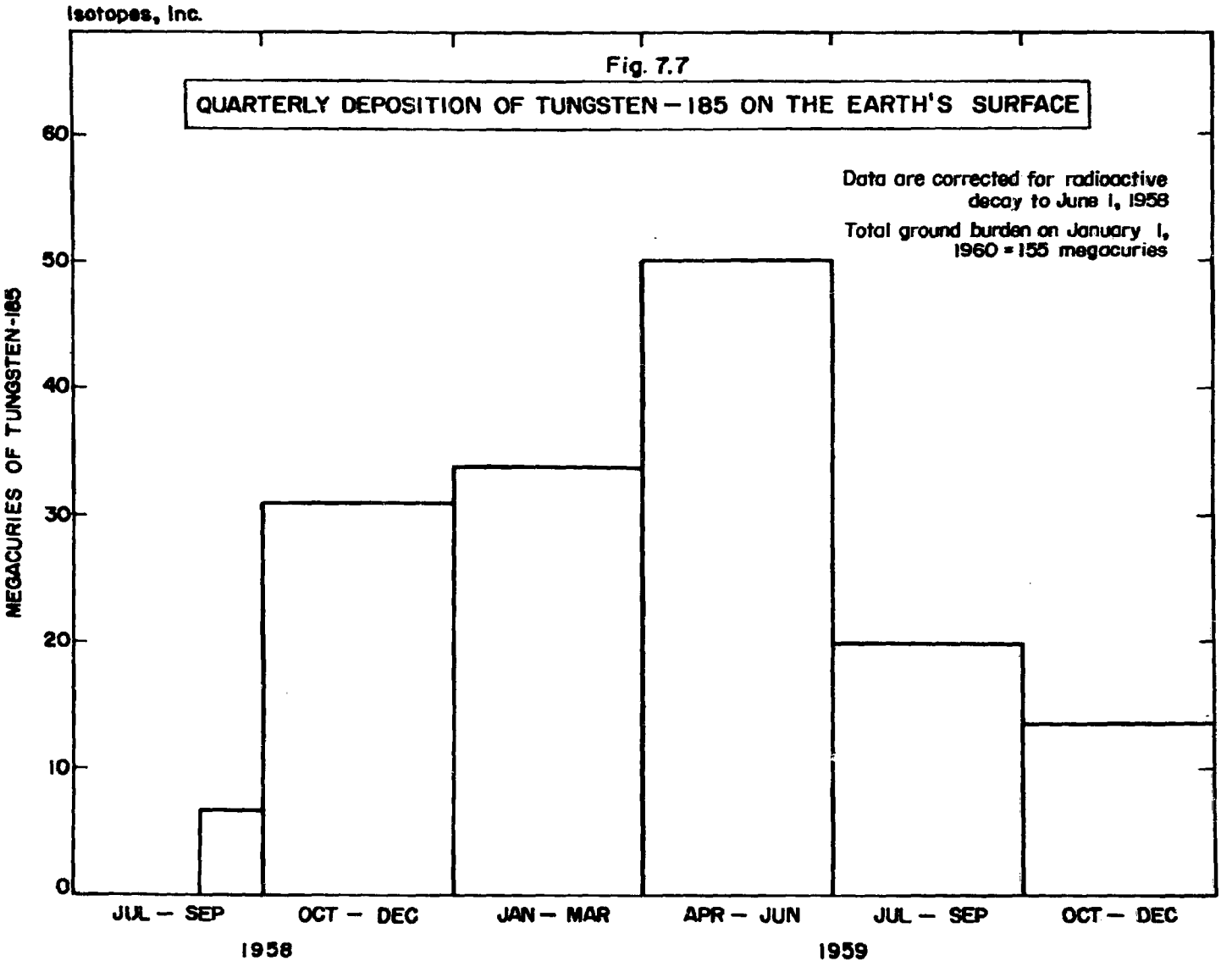
* All concentrations expressed in mc W^{185}/mi^2/inch corrected for radioactive decay to June 1, 1958

** Indicates interpolated values.

Table 7.13. Variation of Tungsten-185 Deposit* on Earth's Surface

Latitude Band	1958		1959			
	Sept.	Oct.-Dec.	Jan.-Mar.	Apr.-June	July-Sept.	Oct.-Dec.
80°-90°N	0.01	0.09	0.06	0.14	0.03	0.02
70°-80°N	0.04	0.38	0.26	0.56	0.13	0.09
60°-70°N	0.16	1.49	1.04	2.19	0.51	0.35
50°-60°N	0.41	3.71	2.57	5.45	1.28	0.88
40°-50°N	0.61	5.50	3.82	8.07	1.90	1.30
30°-40°N	0.63	5.70	3.96	8.38	1.97	1.35
20°-30°N	0.78	1.91	2.78	3.87	1.26	0.77
10°-20°N	0.80	0.80	2.81	2.81	2.70	0.62
0°-10°N	0.87	1.83	2.61	4.82	0.68	1.41
0°-10°S	0.66	1.65	0.85	4.05	2.37	3.32
10°-20°S	0.51	1.74	0.58	3.16	3.12	0.25
20°-30°S	0.11	1.98	1.16	1.97	0.18	1.61
30°-40°S	0.38	1.25	3.36	1.36	1.10	0.48
40°-50°S	0.47	1.54	4.16	1.68	1.36	0.59
50°-60°S	0.33	1.07	2.89	1.16	0.94	0.41
60°-70°S	0.10	0.34	0.91	0.36	0.29	0.13
70°-80°S	---	0.01	0.03	0.01	0.01	---
80°-90°S	---	---	0.01	---	---	---
Total Deposit for Time Interval	6.87	30.99	33.86	50.04	19.84	13.56

*Megacuries of tungsten-185 corrected for radioactive decay to June 1, 1958.


Isotopes, Inc.
Fig. 7.7
QUARTERLY DEPOSITION OF TUNGSTEN – 185 ON THE EARTH'S SURFACE
Data are corrected for radioactive decay to June 1, 1958
Total ground burden on January 1, 1960 = 155 megacuries
MEGACURIES OF TUNGSTEN-185
0
10
20
30
40
50
60
JUL – SEP
OCT – DEC
JAN – MAR
APR – JUN
JUL – SEP
OCT – DEC
1958
1959

on nuclear weapons testing schedules. This conclusion was discussed recently by Walton[15]. Although this general picture of the fallout pattern of tungsten-185 is apparent from the results of some individual observation stations, it is not at all clear for all stations in the 30°N - 60°N band. For example, at Westwood and Pittsburgh this spring-rise is clearly seen in the observations of concentrations in individual rains, while the data from Bedford, as shown recently by Martell[14], do not reflect this increase in any pronounced way, but tend to show no major increase or decrease during this period. On the other hand, the monthly data available for Vermillion, South Dakota and Tulsa, Oklahoma exhibit a decrease in concentration from the first to the second quarter of 1959. From the second to the third quarter of 1959 a decrease is generally observed in the concentration of tungsten-185 in precipitation with only a few exceptions in the 30°N - 60°N band. This decrease also follows the strontium-90 deposition pattern, as the release of material from the stratosphere was greatly reduced after spring 1959. Results from other parts of the globe can hardly be discussed because of the paucity of data. Only an obvious general statement to the effect that concentrations in the Southern Hemisphere appear lower than those in northern latitudes can be made. No seasonal effect is really apparent in the data from the Southern Hemisphere.

Estimates of the yields of tungsten-185 during the Hardtack test series have given values of 108 and 177 megacuries as the total production for a reference date of June 1, 1958. The present calculation of a total ground-level burden of 155 megacuries by December 31, 1959 falls "conveniently" within the above range. Certainly the estimates of the total injection and ground-level burden are exceedingly uncertain and it is surprising that such reasonable agreement is obtained. Calculations of the stratospheric burden of tungsten-185, as discussed in Chapter 6, indicate a loss of one-half of

the original stratospheric injection (HASP estimate--108 megacuries) during the first nine months, i.e., by about March 1959. Alternatively, about 50 megacuries of tungsten-185 were estimated as remaining in the stratosphere at this time. If the higher estimate of the stratospheric injection is accepted, this result suggests a mean residence time which is much shorter than that calculated by HASP--actually $t_{1/2}$ is about 5 months. A third estimate may be determined from the observed ground-level deposit of tungsten-185 at the end of March 1959 and the known stratospheric inventory for the same time. This results in a value for $t_{1/2}$ = 7 months and an initial stratospheric injection of about 122 megacuries referred to June 1, 1958.

A recent article by Hardy[16] has described a similar study of tungsten-185 deposition on the earth's surface. It was concluded that during the period August 1958 through December 1959 a total of 100 megacuries were deposited (reference date - June 1, 1958). The procedure adopted to obtain this figure involved integration on a monthly basis of the average tungsten-185 deposits, expressed in mc/mi^2, for twenty degree latitude bands. No account was taken of the distribution of the sampling sites on the earth's surface with respect to the world-wide pattern of precipitation.

Summary

From the data which are available on the variations of tungsten-185 concentrations in precipitation it is possible to calculate the surface burden of tungsten-185 at different times during 1958 and 1959. It is apparent that the peak deposition occurred in spring 1959, although considerable amounts were

brought down during the late fall and winter months of 1958. This conclusion can probably be related to the levels of tungsten-185 in the lower stratosphere.

Because of large uncertainties in the yields of the weapons containing tungsten-185, material balance studies are themselves very uncertain. It is clear, however, that the "residence half time" of tungsten-185 in the stratosphere, following its injection in 1958, was in the range from 5 to 9 months.

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CHAPTER 8

REMAINING PROBLEMS IN WORLD-WIDE FALLOUT

The High Altitude Sampling Program has contributed substantially to the solution of the major problems in the prediction of the behavior of stratospheric fallout, but none of these problems has been completely solved and significant disagreement remains concerning all. Thus there remains some question as to exactly how much radioactivity actually stablilized in the stratosphere and how much was still there on any particular date. Similarly there is still some disagreement as to the mean residence time of debris in the stratosphere and some debate even on the method of application of the concept of a mean residence time. Very basic differences of opinion remain concerning the relative importance of the roles played in the transfer of debris by the various mechanisms of stratospheric mixing. Discussion has centered especially on the possible existence of an organized meridional circulation within the stratosphere. Finally, there is still insufficient evidence to determine accurately the relative importance of the possible mechanisms of transfer of debris from the stratosphere into the troposphere.

STRATOSPHERIC INJECTIONS AND INVENTORIES

Because direct observations have been made of the height of stabilization of clouds from United States weapon tests and because there is reliable information on the yields of these tests, it is possible to estimate with some accuracy the amount of debris injected into the stratosphere by United States detonations. The only serious uncertainties in these estimates result from our rather limited knowledge of the relative distribution of the radioactive debris from these tests between local and long-range fallout. Nevertheless, since there is little evidence on the distribution of radioactivity within stablized clouds, the vertical distribution of activity within the stratosphere is still in question, and with it the calculation of the total stratospheric burden.

Much less information is available on the amount, and especially on the vertical distribution, of radioactivity injected into the stratosphere by Soviet tests. The United States Atomic Energy Commission has given estimates of yields from Soviet test series which presumably are fairly reliable[1], but the heights of stabilization and stratospheric injections from such tests can only be roughly approximated. The low tropopause in the temperate and polar latitudes, where Soviet tests are performed, tends to maximize the stratospheric component of atmospheric injections, but apparently the temperature structure of the polar atmosphere serves to inhibit the rise through it of the clouds from nuclear detonations. Thus, the HASP sampling consistently encountered clouds from Soviet tests at lower altitudes than would have been expected from data

compiled by the United States Atomic Energy Commission during tests at Eniwetok of weapons of equivalent yield. It is possible that clouds from Soviet tests did rise to relatively great heights above the site of detonation but then subsided to lower altitudes before being sampled by Crowflight aircraft, but there is insufficient evidence of a stratospheric transfer process which could accomplish this.

The Atomic Energy Commission[1] has released tables of total and fission yields for nuclear weapons exploded between 1952 and 1958. About 92 megatons of fission yield, equivalent to about 9 megacuries of strontium-90, were produced by these tests, but not all entered the stratosphere. Libby[2] has estimated the stratospheric injections of strontium-90 and has presented diagrams depicting the trend in the stratospheric burden with time. Similar diagrams have been prepared during the course of HASP[3,4]. These are shown in Figure 8.1.

Libby's estimates were based on known weapon yields and on stratospheric residence times (τ = 5 to 10 years) calculated from the early, rather limited, measurements of surface fallout. The first HASP estimates, given by Stebbins[3], were based on weapon yields and were fitted to a mid-1958 stratospheric burden of 1.0 megacurie calculated from HASP data. The fit was obtained by assuming a stratospheric residence half-time of 12 months for debris injected into the tropical stratosphere and 6 months for debris injected into the non-tropical stratosphere. The stratospheric yields used by Libby were based on the assumption that all debris from megaton weapons, and none from sub-megaton weapons, stabilized in the stratosphere. The yields used in the first HASP estimate were

calculated at Headquarters, DASA, based on the weapon yield-cloud height relationship observed during United States weapon tests and on observed or assumed tropopause heights. For both estimates local fallout was assumed to be negligible from air bursts and about 80 percent from surface land bursts, but Libby assumed 20 percent local fallout from surface water bursts while 70 percent was used in the HASP calculation. Presumably this was a major factor in producing the disparity in estimates of the stratospheric injection by the 1954 Castle series of tests.

The second HASP estimate, prepared by Stebbins[4] at Headquarters, DASA, was based on a more thorough analysis of the dependence of height of stabilization of the radioactive cloud on the conditions of detonation. Curves depicting the dependence of percent stratospheric injection on the location and conditions of detonation were prepared during that analysis and are reproduced here in Chapter 5 (Figure 5.6). Some account was also taken of the variation of stratospheric residence time with altitude, as well as latitude, of injection. Thus, while a residence half time of 5 months was assumed for polar injections, 10 months was used for injections into the lower tropical stratosphere and 20 months for those into the upper tropical stratosphere. Recognition was then made of the uncertainty of scavenging factors to be applied to local fallout and a series of factors was tried. Two extreme sets which gave reasonable fits for the calculated HASP stratospheric burdens (for 1958: 0.95 MC.; early 1959: 0.8 MC.; and late 1959: 0.7 MC.) were: (1) 50% scavenged for land surface and 30% for water surface shots, and (2) 65% scavenged for land surface and 50% for water surface shots.

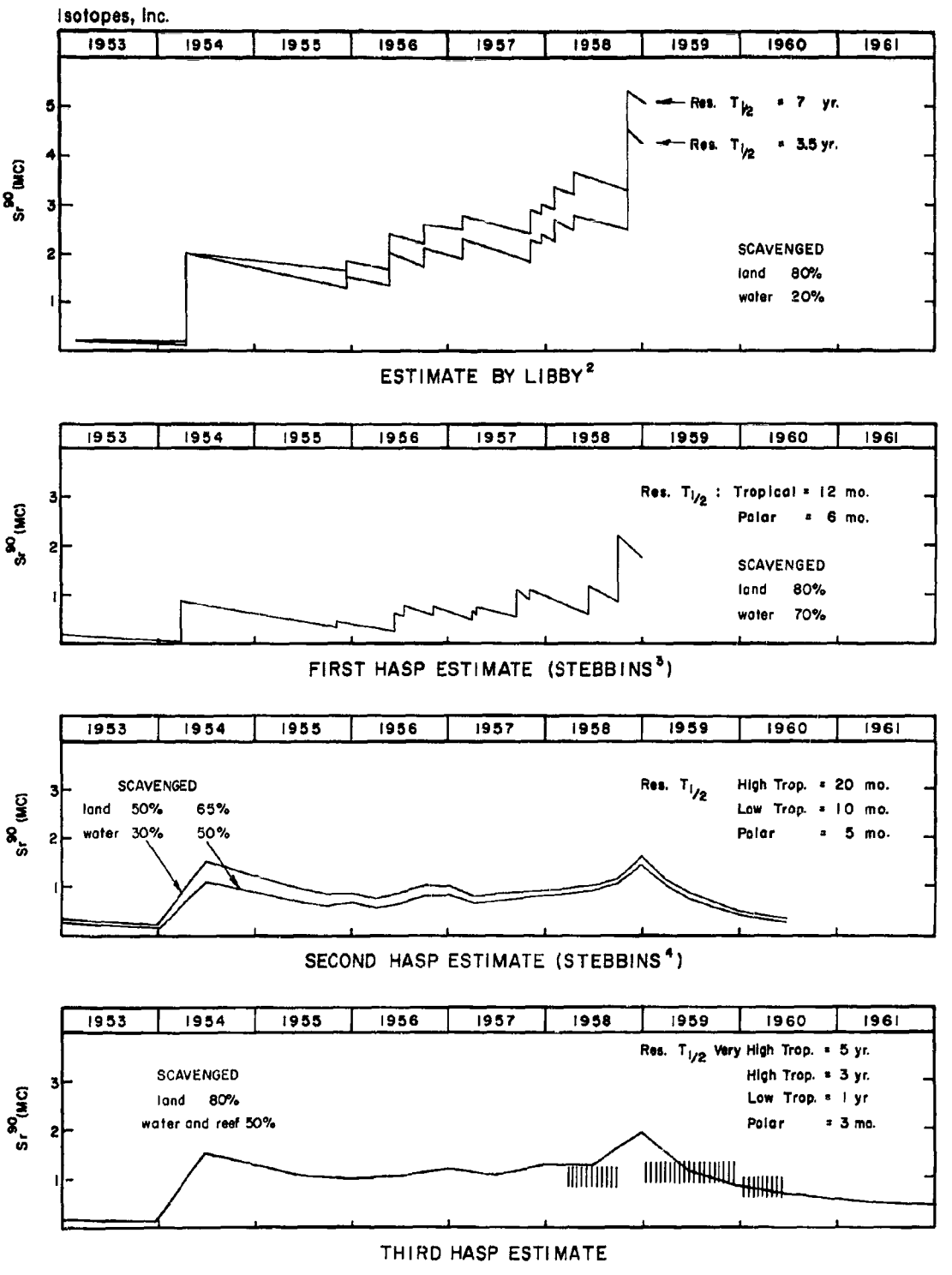

FIG. 8.1 ESTIMATES OF THE TREND WITH TIME OF THE STRONTIUM-90 BURDEN OF THE STRATOSPHERE

In the third HASP estimate we have used scavenging factors of 80% for land surface and 50% for water surface bursts, longer stratospheric residence times for tropical injections and shorter residence times for polar injections in an effort to fit the current HASP estimates of stratospheric burdens (about 1.0 MC. in mid-1958, about 1.1 MC. in mid-1959, and about 0.8 MC. in mid-1960). A residence half-time of 3 months was assumed for polar injections, 12 months for tropical injections below 70,000 feet, 36 months for those between 70,000 and 90,000 feet, and 60 months for those above 90,000 feet. As in the earlier estimates, the contribution from the high altitude rocket shots, Teak and Orange, was excluded.

Other measurements of strontium-90 in the stratosphere have indicated concentrations similar to those found during HASP. Machta and List[5] used data from Project Ash Can balloon sampling to calculate stratospheric burdens of about 1.2 megacuries of strontium-90 during 1957 and 0.8 megacurie during 1958, though they were not completely convinced of the reliability of these data. List and Telegadas[6] used data from filter sampling of the atmosphere by B-57 and U-2 aircraft to calculate a stratospheric burden of about 1.0 megacurie of strontium-90 in May 1960. This later estimate, like the HASP estimate of about 0.8 (or about 1.0 if debris below 70,000 feet attributed to the rocket shots was not excluded) for the same interval, evidently did not include that portion of debris from the high altitude rocket shots which still remained in the mesosphere or high stratosphere.

As should be expected, the various HASP estimates of the stratospheric burden during 1958 have more or less agreed with each other, since they were

all based on the same data. However, burdens calculated for later years changed, and presumably improved, as results from measurements of stratospheric concentrations during 1959 and 1960 also became available. Because the average stratospheric residence times used in the HASP calculations were much shorter than those used by Libby, the HASP estimates of the burden during mid-1958 are only about one half to one third that of Libby. Several factors caused this discrepancy. Libby used data on weapon yields to calculate stratospheric input and data from surface measurements to calculate fallout. His assumption of little local fallout from surface water bursts leads to higher calculated injections by United States test series than were indicated by the HASP calculations. At the same time, Libby assumed less deposition of stratospheric fallout than did HASP because of his identification of the excess strontium-90 deposited in north temperate latitudes as intermediate fallout from Nevada tests. Though Libby's original estimates of the deposition rate of world-wide fallout were shown to be too low by measurements made during 1959 especially, the differing estimates of the total stratospheric injection have not been resolved, and speculations persist[7,8] that large quantities of debris from high yield surface detonations linger in the upper atmosphere, above the reach of aircraft or balloon sampling. However, such speculations appear to ignore the observed relatively rapid downward movement of debris deposited originally in the mesosphere by the 1958 rocket shots, as is discussed below.

THE STRATOSPHERIC RESIDENCE TIME

The original concept of detention of nuclear debris injected into the stratosphere, followed by slow diffusion into the troposphere and fallout to the ground, was devised by Libby[9]. He assumed very rapid mixing of debris in the stratosphere followed by loss of debris through the tropopause according to first order kinetics. This concept is quite convenient, for it lends itself readily to the calculation of stratospheric burdens from fallout rates or to the estimation of future fallout rates from known burdens.

Unfortunately, as fallout data accumulated, it became evident that the actual processes were too complicated to be described accurately by simple equations. Thus, Stewart et al[10] concluded in 1957 that stratospheric fallout, for meteorological reasons, had been fed down slowly into the lower stratosphere in a periodic manner, that deposition had taken place preferentially in the middle latitudes, and that deposition in the Northern Hemisphere had been greater than that in the Southern Hemisphere. These conclusions have since been well substantiated. Stewart et al, and Machta[11] believed that the periodicity of fallout rate and maximization of deposition in the north temperate latitudes applied to debris injected by both equatorial and high latitude tests. Martell[12], however, on the basis of surface measurements of fallout, attributed both phenomena primarily to the Soviet test schedule. Spar[13], in discussing the HASP fallout model, predicted more rapid fallout of Soviet debris on the basis of meteorological considerations. Subsequent work, especially measurements of tungsten-185 in precipitation reported by Walton[14] and others, has shown that

Stewart et al were essentially correct, and that debris from United States tests is deposited preferentially during the spring season and in the north temperate zone. However, the very rapid fallout, in the north temperate zone, of debris from Soviet tests has also been confirmed, both by measurements of surface fallout and by HASP data.

In recognition of the differences in behavior between fallout from injections at different latitudes, interpretation of fallout rates from HASP data has included the somewhat arbitrary assignment of different residence times to debris from polar injections, to debris injected into the lower tropical stratosphere and to debris injected into the higher tropical stratosphere. Although this modification of the fallout calculation gives a more reasonable picture of the actual behavior of debris than did the original simple concept, even it is only a rough approximation of the actual situation, for no account is taken of the strong seasonal fluctuations in stratospheric mixing and transfer processes. Nevertheless, this appears to be the best simple approach to the prediction of stratospheric residence times, at least as long as turbulent diffusion is viewed as the primary cause of stratospheric transfer of debris. A fairly good approach to the true rates could probably be obtained if sufficient data were available to assess definitively the effect of altitude, as well as latitude, of injection on residence time, for this appears to be an extremely important factor.

Proponents of an organized meridional circulation as the chief mechanism in the transfer of debris within the stratosphere have not seemed to have any difficulty in reconciling this mechanism with an apparent fallout of debris more or less according to first order kinetics[7, 10]. It would seem, however, that if

turbulent diffusion were subordinate to such organized transfer, that stratospheric fallout would reach the troposphere and the ground in a series of pulses, spaced according to the times of injection and the speed of the circulation. Thus, if the mean stratospheric residence time of equatorial debris were five years, as assumed by Libby, a circulation such as that propounded by Libby and Palmer[7] should cause the highest rate of strontium-90 fallout from an equatorial test series to occur during the fifth year after its injection. Of course, turbulent mixing of debris from each test with uncontaminated stratospheric air would cause some fallout to occur before and some to occur after the mean, but presumably the stratosphere would be cleansed of old debris much more rapidly by an organized circulation than by diffusive mixing. The significance of the "mean" residence time depends quite strongly upon which mechanism is referred to.

In summary we may state that there is now general agreement that there is no single stratospheric residence time applicable to all debris. It is widely recognized that debris from Soviet Arctic injections displays a residence time of only a few months while debris from United States and United Kingdom tests has a longer residence time. The HASP measurements indicate that debris from these latter tests experiences residence times ranging from a few months, for material injected just above the tropopause, to several years, for material injected in or above the mesosphere, and there is no strong evidence which contradicts this.

MECHANISMS OF STRATOSPHERIC TRANSFER

The various postulated mechanisms of stratospheric transfer have been discussed at length in Chapter 6 of this report. Here we will merely review briefly some of the work which has produced results which bear upon the selection of the most likely transfer mechanism.

Brewer[15], to explain some measurements of frost-points in the lower stratosphere over England, hypothesized an organized meridional circulation which carried air poleward from the region of the cold tropical tropopause. Dobson[16] used a modification of this concept to explain the seasonal variation in ozone "fallout" rates. Stewart et al.[10] believed that the "Brewer-Dobson" circulation could explain the seasonal variations in the rate of fallout of radioactive debris from the stratosphere, as mentioned earlier in this chapter. Hagemann et al.[17] found anomalously low concentrations of bomb-produced carbon-14 in the lower tropical stratosphere and concluded that an organized meridional circulation could best explain the rapid depletion of bomb debris from this region. Burton and Stewart[18] found evidence for a meridional circulation in measurements they made of stratospheric concentrations of radon daughter products. Libby and Palmer[7] and Storebø[19] used surface fallout data to support different hypothesized circulations.

In spite of the wide support which has been given the theory of an organized meridional circulation, the HASP data on stratospheric concentrations of radioactivity and their change with time have always appeared to be more easily explained if turbulent diffusion is the primary mechanism of stratospheric transfer.

The observed tungsten-185 concentrations especially have appeared to be explicable only in terms of turbulent diffusion, with a meridional circulation playing only a secondary role, if any. Thus, it is of interest to consider briefly the points of disagreement of the two mechanisms of transfer which have been proposed. These points are the hypothesized organized up-welling of air through (or only above[7]) the tropical tropopause, the organized subsidence of air in the polar or temperate latitudes, and the net poleward drift of air in the high stratosphere[10] (and perhaps an equatorward drift of air in the lower stratosphere[7]).

An upward flow of air in the tropical stratosphere is a basic component of the meridional circulations described by Brewer and Dobson, by Libby and Palmer, and by Storebø. Evidence for such a flow has been found in the low frost-point of air in the non-tropical stratosphere (indicating passage of this air through the tropical tropopause region[15]), in the concentrations of ozone in the tropical stratosphere which are anomalously low and which display a maximum at anomalously high altitudes[20], and observed concentrations of bomb-generated carbon-14[17], which were found to be quite small in the lower tropical stratosphere in spite of the injection of debris into that region by United States and United Kingdom tests. A number of determinations[21,22] have now been made of frost-points in the stratosphere at altitudes higher than those sampled by the British and interpreted by Brewer, and it is fairly well established that the frost-point, after passing through a minimum in the lower stratosphere, increases again at higher altitudes. The reason for this phenomenon is not known, but it certainly is not readily explained in terms of an organized upward flow of air in the tropical stratosphere. Similarly the air which Hagemann et al. found to

contain low concentrations of carbon-14, nevertheless, contained fairly high concentrations of fission products, as indicated by Project Ashcan samples[23] collected by the same balloons which collected the gas samples. Neither have HASP measurements indicated any general tendency for rapid depletion of the fission product concentrations in the air of the tropical stratosphere. The high activities which characterized the lower tropical stratosphere during and immediately after series of tests in the equatorial regions did decrease rapidly, both by mixing with uncontaminated air and by fallout, but concentrations leveled out within a few months and subsequent decreases were slow and were seasonal in character, probably because of seasonal variations in stratospheric mixing rates. As has been mentioned above, the HASP tungsten-185 data appear to refute completely any organized upward flow of air from the lower 15,000 feet of the tropical stratosphere. Thus, the only remaining tracer evidence which suggests upward flow, the vertical concentration profile of ozone in the equatorial stratosphere, is contradicted by several other lines of evidence.

There is more basis for the assumption of subsidence of stratospheric air in the polar and temperate latitudes. As has been mentioned above, Brewer[15] considered the maintenance of low frost-points in the low polar stratosphere to be an indication of an organized downward flow of air through that region. Dobson[16], Paetzold[24], Brewer[25] and others have attributed the observed inflow of ozone into the polar stratosphere during the late winter and spring to such an organized downward flow. Stewart et al[10] attributed the spring increase in fallout concentration in the lower polar stratosphere and the troposphere to the same mechanism, and Martell[12] used it to explain the short stratospheric

residence time of polar injections. This apparent downward flow of air during the polar winter has been related by Libby and Palmer[7], and by others, to disturbances in the winter polar vortex, specifically to the explosive warmings in the stratosphere which have been observed during the Northern Hemisphere winters. During HASP[26] the transfer of tungsten-185, strontium-90, ozone and other "tracers" into the lower polar stratosphere has been attributed predominantly to turbulent diffusion ("Austausch"), which apparently has been most effective during the winter months. This seasonal acceleration in mixing rates may be due to the high wind speeds which characterize the winter polar vortex. However, Kalkstein[27] attributes it to the breakdown of the vortex, not to the properties of the stable vortex. Most of the changes in the stratospheric distribution of nuclear debris which were observed to occur during HASP did not seem to result from sudden major shifts in air masses, but rather from longer range shifts occurring over the course of several months. In a few instances, changes did appear to occur suddenly, between two sampling missions, but often such missions were flown several weeks apart. Quite often, of course, large variations in concentration were observed from mission to mission, even during the moratorium on testing, but these changes were normally ephemeral. Moreover a comparison of Project Ashcan Ce^{144}/Sr^{90} measurements[28] with those made during HASP shows that a year was required for debris from the rocket shots to descend from 90,000 feet (in early 1959) to below 70,000 feet (in early 1960). Both the strontium-90 and tungsten-185 from tropical injections appeared to subside to lower altitudes as they mixed poleward, and this subsidence seemed to be typical of the mixing proccesses throughout

the year, not just during a few intervals that might be correlated with explosive warmings of the stratosphere. Certainly it did not seem to fit the pattern expected of an organized meridional transfer[26]. Nevertheless there are characteristics of the distribution in the polar stratosphere which are difficult to explain by mixing processes alone. Most important of these was the virtually complete loss from the stratosphere of debris from the October 1958 Soviet injections in less than a year (see Chapter 6). This phenomenon is most easily explained if some organized downward flow of polar stratospheric air occurs. The stratospheric distribution and concentration of beryllium-7 and phosphorus-32 found in HASP samples seem to argue against such a downward flow, however, so that a contradiction results which is not easily resolved.

The third aspect of stratospheric transfer in which disagreement remains is the relative importance of organized and unorganized meridional movement of stratospheric air. The dew-point measurements reported by Brewer[15] and two of the three results reported by Mastenbrook and Dinger[21] seem to require a net poleward movement of air from the lower tropical stratosphere, virtually undiluted by air with a higher moisture mixing ratio. Similarly, large quantities of ozone are carried into the polar stratosphere during the winter and spring months creating a larger reservoir of ozone there than is present in the equatorial region of formation. Brewer[15], Dobson[16], Paetzold[24], Palmer[7] and others have interpreted such meridional transfer as an indication of an ordered meridional circulation, but Sterebϕ[19], Dütsch[20] and Spar[29] have believed that the ozone transfer at least could be effected by turbulent diffusion. The frost-point data of Mastenbrook and Dinger suggest that different profiles may be

found at the same latitude but different longitudes. Measurements in the Chesapeake Bay area (like Brewer's measurements over the British Isles) indicated a profile which should be characteristic of the tropical stratosphere, but measurements in the Denver area gave quite a different result, a profile which lacked a pronounced minimum immediately above the tropopause. Perhaps meandering streams exist in the stratosphere, carrying parcels of air poleward from the tropical region and other parcels equatorward from the polar region. These streams would form one component of a large scale turbulent mixing which would move large quantities of air (and their contained tracers) through great distances before they became dispersed by small scale mixing. The existence of such streams is certainly suggested by many HASP measurements. Clouds from Soviet injections at 70° North were often intercepted by Crowflight aircraft at 30° North rather than at higher latitudes. Similarly, wide variations in concentrations of tracer elements, such as tungsten-185, were found in samples collected in the same region only a few days apart. These suggest that stratospheric mixing is brought about to a large extent by sporadic shifts in the north-south component of the zonal circulation. More definitive information on these mixing processes could be obtained from the study of tracers originating in the polar stratosphere. The Brewer - Dobson theory would predict negligible movement of such tracers into the tropical region, the Libby-Palmer theory would predict an organized transfer into that region, and the HASP model would predict a rapid equatorward mixing. The beryllium-7 and phosphorus-32 measurements made during HASP appear to be compatible with the HASP prediction, but further evidence would be most desirable.

The meridional transfer processes active in the stratosphere are evidently quite effective in moving nuclear debris across the equator in the stratosphere, though movement of debris across the equator in the troposphere is impeded, as indicated by Lockhart's measurements[30, 31] of fission products in tropospheric air during 1959. The effectiveness of transfer across the equator is demonstrated by the steady fallout of stratospheric debris in the Southern Hemisphere in spite of the fact that almost all stratospheric injections have occurred in the Northern Hemisphere. It appears fairly clear now that more than half of the radioactivity injected by United States tests at 11° North and all of that injected by Soviet tests at high latitudes does fall out in the Northern Hemisphere, but perhaps half of that injected into the equatorial stratosphere by British tests and perhaps one third to one quarter of the United States injections falls out in the Southern Hemisphere. HASP data suggest that transfer across the equator from the Northern into the Southern Hemisphere is accelerated during the winter season in the Southern Hemisphere. Depletion of equatorial concentrations was evident during that season as well as during the winter of the Northern Hemisphere, and influx of equatorial debris into the Southern Hemisphere was quite marked during the Southern Hemisphere winter of 1959. The poleward flow of debris from the equator during the winter season was also observed by Murayama and Machta[32] in data from Project Ash Can balloon sampling and in stratospheric carbon-14 measurements from both the Northern and Southern Hemisphere. The rapid lateral spread and fallout of Hardtack debris injected immediately above the tropical tropopause was observed during HASP sampling. Perhaps the virtual equipartition of Hardtack fallout between the Northern and Southern

Hemispheres observed by Lockhart et al[33] is attributable to the spread of debris across the equator in these lowest layers of the stratosphere rather than to its spread in the troposphere. This might explain the easy passage of Hardtack debris across the equator while Soviet debris was restricted to the Northern Hemisphere[30].

MECHANISMS OF TRANSFER TO THE TROPOSPHERE

The various possible mechanisms of transfer of radioactive debris from the stratosphere into the troposphere were discussed in Chapter 6. HASP data have not been of such a nature as to shed very much light on the relative importance of these various mechanisms but, combined with the results of other experimental programs, they do indicate the probable importance of at least one of these mechanisms, transfer through the tropopause gap by meandering streams of air.

The low fallout rates observed in tropical areas, even in those with high rates of rainfall, suggest that little loss of debris from the stratosphere occurs through the tropical tropopause. This is quite reasonable in light of the stable character of the tropopause layer at low latitudes. It would appear that debris, except perhaps for that injected into the tropical tropopause layer itself, must be transferred into the polar stratosphere before it can escape to the troposphere. Once it has reached the lower polar stratosphere there are several methods, discussed in Chapter 6, by which such escape may occur. One such method, transfer through the tropopause gap by meandering streams of air, is indicated

by the common occurrence in the stratosphere, at about the altitude of the gap region, of parcels of air of tropospheric origin. These may be identified by low concentrations of radioactive debris. Many examples may be found in HASP flight cross sections (see Chapter 4, Figures 4.101, 4.104, 4.128, 4.129, etc). Similar evidence of tropospheric air entering the stratosphere in this region is found in the common minimum in the vertical profile of ozone concentrations[24,25]. It seems unlikely that large parcels of undiluted tropospheric air could enter the stratosphere by any other normal mixing process. Evidence for the transfer of some stratospheric debris out through the gap region may be found in Gustafson's[34] detection of rhodium-102 in tropospheric air in late 1959, before it had mixed into the lower layers of the polar stratosphere. Much of the observed variation of concentrations of radioactive debris and of ozone in the polar stratosphere between 45,000 and 60,000 feet could be explained by alternate mixing of air in that region first with air in the lower tropical stratosphere and then with air in the upper tropical troposphere. Such mixing would evidently consist of interchange of parcels of air between these regions by meridional meandering of predominantly zonal currents. Such a phenomon is not at all unlikely.

SUMMARY

In summary we may again state that, though the HASP data have not completely resolved the uncertainties in the interpretation of fallout data, they have contributed a large body of evidence which may be used to reduce greatly almost all of these uncertainties. The uncertainties which remain in the fundamental parameters used to describe the behavior of fallout can be further reduced by future measurements of fallout, both by means of ground level collections and by airplane and balloon sampling. Study of the data which have already been collected should also be continued, for the full measure of potentially useful information they contain has yet to be extracted from them.

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CHAPTER 9

THE HAZARDS FROM RADIOACTIVE FALLOUT

The biological hazard which results from the deposition of radioactive materials both on the earth's surface and in the human body after the detonation of nuclear devices is an extremely controversial subject. It has received attention not only because of the scientific problems involved, but also because of the political and humanitarian implications. The extent of this biological hazard is the question of most importance at the present time. During the past six or seven years, since the testing of nuclear weapons resulted in the deposition of large quantities of fallout on the surface of the earth, this problem has received increasing study by scientists and statesmen alike. Its extent and what the future consequences of such cumulative dosages and dose rates will be remain as unsolved problems and subjects of considerable debate by informed and uninformed sources alike.

A prerequisite to any discussion of the hazards from fallout radioactivity is a knowledge of the level of radiation to which the human being is subjected. It is in this phase of the problem that HASP has much to contribute. Not only do the results of the HASP program allow us to calculate the amount of radioactivity remaining in the stratosphere at a given time, but they permit future concentrations

of radioactivity on the ground to be assessed from the rate at which the current levels of stratospheric material are expected to be deposited. Data[1] from HASP have been used in the past to delineate the rates of fallout deposition. It was demonstrated that the original theories of slow fallout, with residence times in the stratosphere of up to ten years, are not tenable. Consequently biologists have been compelled to consider the possible hazards from the short-lived products of nuclear detonations in addition to the hazards from long-lived nuclides such as strontium-90 and cesium-137, which have always been given much attention. It was not until 1959 that the subject of the short-lived nuclides was given the study which it merited. Of course, these nuclides become important only during periods of weapons testing or nuclear war and with the current moratorium, which has been in existence for almost three years, we are not confronted with this problem. Nevertheless it is interesting to compare the acute dose from the short-lived nuclides with the chronic irradiation from the bone seekers.

Our knowledge of the chronic effects of irradiation from nuclides present in the body at extremely low concentrations remains limited, but recently considerable emphasis has been placed on studies which can possibly provide information on this subject. One of the most obvious methods for elucidating these effects is to study populations exposed to elevated levels of natural radioactivity and compare the observed genetic and somatic effects with the effects in populations living in areas of lower background radiation. Both statistical and technological problems as well as human problems are involved in this type of study. The evaluation of the sampling techniques and the significance of the observations which might be made in areas of higher natural radioactivity than normal is always problematical. Furthermore, there is the practical problem of accurate measurement

of natural activity levels.

Finally we have the problem of determining the magnitude of the hazards associated with the peaceful applications of radioactivity and ionizing radiation. Included in this category is the widespread diagnostic use of X-rays and other applications, together with the use of high energy gamma sources and the expanding usage of radioisotopes, produced generally as by-products of atomic pile operations, in the physical and biological sciences.

In this report we are concerned only with the potential hazards resulting from nuclear weapons testing. We believe it is pertinent, however, that other sources of ionizing radiation and the activity levels to which human beings are subject, be listed alongside those levels which result from radiation produced by weapons testing. Only the levels of activity are reported in this chapter and the reader is referred to more detailed treatises of the effect of this irradiation on the human body.

The relative hazard presented by a particular radioactive nuclide is dependent upon several factors, which include

1. the amount of the nuclide ingested or inhaled into the body,
2. the residence time of the nuclide within critical parts of the body,
3. the type and rate of nuclear disintegration which the nuclide undergoes.

In this chapter these factors are considered for several individual nuclides. A review is presented of the current intake levels of strontium-90, cesium-137, plutonium, carbon-14 and the projected levels of activity of these nuclides in the body and the environment. From these data it is possible to make some calculations of the dosages and dose rates from these nuclides. In addition

the problem of the radiation from short-lived products of nuclear testing is discussed briefly and a final comparison is made of the dosages from natural and fallout radioactivity.

RADIATION HAZARD FROM STRONTIUM-90

From the inception of nuclear testing it was evident that strontium-90 was a potentially hazardous nuclide. It is produced in relatively large amounts during fission, the half life is 28 years, and because of its chemical similarity to calcium it is incorporated and retained in bone. It is currently believed that bone should be considered as the "critical deposition site" and continued irradiation of this part of the body could lead to bone cancer or leukemia. The concentration of strontium-90 in bone is ultimately governed by the concentrations of strontium-90 in the diet and the discrimination factors involved in the deposition process.

Strontium-90 in Diets

The majority of research conducted to determine the strontium-90 concentrations in human diets has been restricted to Western cultures. Relatively little work has been performed in rice-diet areas, although the concentrations of strontium-90 in the bones of people residing in these areas have attracted some attention. It is generally believed that the amount of strontium-90 absorbed by bones is governed by the strontium-90 to calcium ratio in the diet. Since the major sources of calcium in the diet of Western culture are dairy products, it follows that, if the strontium-90 to calcium ratios in the other contributors to the diet do not vary over an extremely wide range, the

strontium-90 to calcium ratio in the diet will be governed largely by the concentration in dairy products. For this reason milk was chosen as a material which could be used to monitor the dietary intake of strontium-90. Recently, however, more attention has been given to the strontium-90 contents of total diets to estimate more accurately the strontium-90 intake.

Table 9. 1 shows the strontium-90 content in the diet of various countries during different periods of 1959, and Table 9. 2 lists the average strontium-90 concentration in diets from New York City, San Francisco, and Chicago during early 1960. The lower strontium-90 concentration observed in the San Francisco diet in 1960, compared to the other two United States cities, is not unexpected because of the lower deposits of this nuclide in the drier areas of the United States. New York City has not experienced any sharp decline in the amount of radioactivity in food since nuclear testing ceased in late 1958. Previous results obtained by HASL[9] and Consumer Reports[6] for the New York area are listed in Table 9. 3 to indicate the variations in strontium-90 concentrations observed during the years 1958-1960. Only the results of Consumer Reports[6] were obtained on actual diet samples. The other data were determined from the analyses of individual foods in an average diet. Past investigations of the factors controlling the concentration of strontium-90 in the diet led to the conclusion that a considerable fraction of the radioactivity present in foods had been deposited directly on plants, without passing through the soil. Thus Burton et al[10] demonstrated that in 1958, 80 percent of the strontium-90 in United Kingdom milk was derived directly through "foliar uptake" and only 20 percent was derived from the cumulative amounts of strontium-90 present in

Table 9.1 Average Strontium-90 Content in Diets of Several Areas During 1959

Country	Strontium-90 Intake ($\mu\mu c\ Sr^{90}/g\ Ca$)
United Kingdom[2]	9.0
Germany[3]	11.5
Japan[4]	13.4
United States, Cincinnati[5]	14.0
United States, 25 Cities[6] (Nov)	11.8
Ecuador[7]	2.5
Peru[7]	2.6
Vietnam[7]	9.4

Table 9.2 Average Strontium-90 Content in Diets of Three U. S. Cities During Early 1960[8]

City	Strontium-90 Intake ($\mu\mu c\ Sr^{90}/g\ Ca$)
New York (March)	11.4
Chicago (May)	11.1
San Francisco (March)	5.3

Table 9.3 Estimates of Strontium-90 in Diet for New York City (1958-1960),[9] Tokyo (1957-1960)[4] and Kagoshima (1957-1960)[4]

Time			Strontium-90 Intake μμc Sr^{90}/g Ca
(a) New York			
	1958	HASL	12.6
August	1959	HASL	14.0
November	1959	HASL	17.7
November	1959	Consumers Union[6]	12.4
December	1959	HASL	12.4
March	1960	HASL	11.4
(b) Tokyo			
June	1957		3.6
November	1958		6.8
June	1959		10.9
September	1959		13.7
December	1959		14.4
March	1960		21.9
(c) Kagoshima			
August	1957		0.74
November	1957		153.0
February	1958		2.82
May	1958		1.00
August	1958		0.84
November	1958		8.5
June	1959		12.0
September	1959		11.1
December	1959		12.1
March	1960		20.1

the soil. Initial evidence for this conclusion was derived from the observation that the strontium-89 to strontium-90 ratio in milk in 1958 in the United Kingdom[10] was approximately one half of the ratio in the current deposition in rainfall and about five times higher than in the cumulative deposits of these nuclides in soil. It was therefore apparent that the concentrations of strontium-90 in milk were highly dependent on the amounts of strontium-90 recently deposited. Field experiments involving the application of a fine spray of strontium-89 to soils and crops[10] confirmed that a considerable fraction of the radioactivity in the crops was derived from the radioactivity which had been directly deposited on the surface of the plant and retained through "foliar uptake," rather than on that which passed through the soil. Of course, the relative importance of foliar uptake and uptake through the roots will obviously vary according to the rate of deposition of activity at a given time. In the spring of 1960, when the rate of fallout was approximately an order of magnitude less than the rate during spring of 1959, it was expected that a considerable reduction from the 1959 values would have occurred in the strontium-90 content of milk, and probably in the total diet also. This decrease is not yet apparent in the data shown in Table 9.3 for the New York and Japanese cities. More recent data[23], however, on strontium-90 in United States milk during the second quarter of 1960 indicate that the concentrations have dropped by a factor of two from the previous year. Thus at least half of the strontium-90 in milk must be the result of direct absorption.

The actual intake of strontium-90/day may be calculated from the data in Table 9.2, assuming a calcium intake of 1 g/day for people in the United

States, and approximately 0.6 g/day for the Japanese population. Thus the Japanese diet results in a strontium-90 intake in 1959 which is close to one half of the amount in the average United States diet.

Concentrations of strontium-90 in bone are, of course, related to the nuclide's concentrations in the diet and can be calculated for any individual at any time if the following factors are known:

a. the discrimination factor between strontium and calcium in passing from diet to bone;

b. the average weight of calcium being added to the skeleton;

c. the average concentration of strontium-90 in the individual's diet during the history of the individual;

d. the average rate of growth, exchange, and replacement of bone;

e. the discrimination factor between strontium and calcium from the mother's diet to the fetus.

Most of these factors are fairly well known, with the exception that future strontium-90 concentrations in the diet are somewhat uncertain.

Future Concentrations of Strontium-90 in the Diet

Estimates of future concentrations in diets can be calculated if certain assumptions are made concerning the relative contributions of strontium-90 from the two routes of entry into foodstuffs. The work of Burton et al[10], indicates that the nuclide concentration in the diet is greatly dependent on the rate of fallout in a given year. It is not yet certain whether the data of Burton et al[10] are applicable to diets from other parts of the world and for this reason we have chosen to calculate future diet concentrations on the basis of several different combinations of cumulative and rate factors. As a

starting point a dietary level in the 30°N-50°N latitude band of 15 $\mu\mu c\ Sr^{90}/g\ Ca$ was assumed for 1959. Concentrations for other years were calculated by substituting the appropriate data in the following equation:

$$C = a(F + 1/2\, f) + b\, f\ ,$$

where

C = concentration of strontium-90 in diet ($\mu\mu c\ Sr^{90}/g\ Ca$),

F = cumulative deposit of strontium-90 at beginning of year ($mc\ Sr^{90}/mi^2$),

f = fallout deposited during the year in question (mc/mi^2),

a and b = proportionality constants ($\frac{\mu\mu c\ Sr^{90}/g\ Ca}{mc\ Sr^{90}/mi^2}$).

Thus

a(F + 1/2 f) = cumulative factor, involving soil strontium-90, and

b f = rate factor, involving strontium-90 from fresh fallout.

For various fractional contributions from the cumulative and rate factors, different values were calculated for a and b and were then applied to determine C for other years. Values of F and f in the 30°N-50°N latitude band for the period 1955-59 were taken from the data given in Chapter 7, and for the years after 1959 the results were calculated from Figure 7.5, assuming that the fraction of the total fallout deposited in the 30°N-50°N band was the same as in 1959. Values of F and f are shown in Table 9.4 and the predicted diet concentrations are shown in Figure 9.1. Before going into discussion of the implications of these predictions as far as bone concentrations are concerned a brief review of investigations to date on strontium-90 in bone is presented.

Table 9.4. Average Values of Fallout Deposit and Deposition Rates in the 30°N - 50°N Latitude at Different Times

Date	Sr^{90} Deposit (mc/mi^2)	Interval	Sr^{90} Annual Increment (mc/mi^2)
1 July 1954	(1.5)		
		1954-1955	4.7
1 July 1955	6.1		
		1955-1956	4.0
1 July 1956	9.9		
		1956-1957	6.0
1 July 1957	15.6		
		1957-1958	12.7
1 July 1958	27.8		
		1958-1959	16.7
1 July 1959	43.6		
		1959-1960	4.4
1 July 1960*	50.4		
		1960-1961	2.1
1 July 1961	51.3		
		1961-1962	1.3
1 July 1962	51.3		
		1962-1963	0.8
1 July 1963	50.9		
		1963-1964	0.7
1 July 1964	50.4		
		1964-1965	0.5
1 July 1965	49.6		
		1965-1966	0.4
1 July 1966	48.8		
		1966-1967	0.3
1 July 1967	47.9		
		1967-1968	0.2
1 July 1968	46.9		
		1968-1969	0.2
1 July 1969	46.0		
		1969-1970	0.1
1 July 1970	45.0		

* For 1960 and following years the strontium-90 deposit and annual increments predicted on basis of data given in Chapter 7.

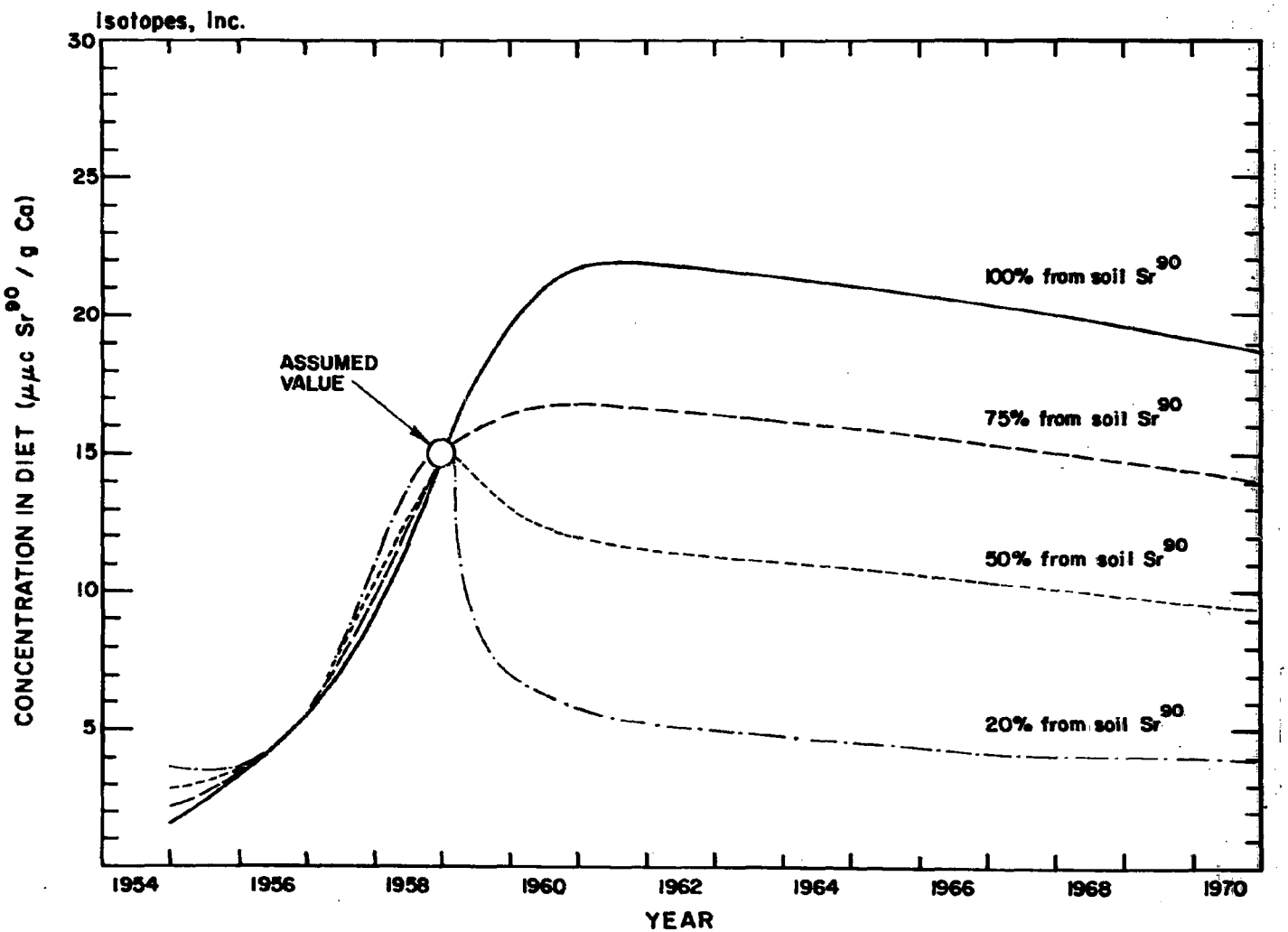


FIG. 9.1 PREDICTED CONCENTRATIONS OF STRONTIUM-90 IN THE DIET, 30°N-50°N (SLIGHTLY IDEALIZED)

Strontium-90 Concentrations in Bone

Several studies of strontium-90 concentrations in bone have been conducted since 1954, but undoubtedly the most comprehensive work was performed by Kulp and his co-workers[11, 12, 13, 14]. In this program over 9000 samples of bone were analyzed with the object of determining:

1. the world-wide geographical variations of strontium-90 concentrations,
2. the age effect on strontium-90 concentrations in bone,
3. the relative concentrations in different bones and
4. the probable future concentrations.

The latest report[14] from this group served to elucidate practically all these factors although some uncertainty remains concerning future concentrations of strontium-90 in bones.

Past and Current Concentrations in Bone

Since 1954 strontium-90 concentrations in bones have shown a gradual increase. Although some of the data in the earlier work do not exhibit this increase very clearly, the results from 1958 and 1959 demonstrate this rise very distinctly. As an illustration of this regular increase, Table 9.5 gives the results of strontium-90 analyses on New York City adult cadavers and some samples from the United Kingdom[15]. These yearly averages for adults are directly comparable since it was demonstrated that the average strontium-90 concentration in adults for a given year was essentially independent of the age of the individual. Of course, adult bones possessed the minimum concentrations of strontium-90 at this time because they incorporated the radioactivity mainly by exchange processes, whereas

Table 9.5 Variation with Time of the Strontium-90 Concentrations in Human Bones from New York City[14] and the U. K.[15]

New York Skeletons[14]

Year	Number of Samples	Ave. Sr^{90} Conc. ($\mu\mu c$ Sr^{90}/g Ca)	Standard Deviation	Standard Error on Mean
1953	2	< 0.005		
1954	26	0.007	± 0.009	± 0.002
1955	58	0.027	± 0.020	± 0.003
1956	131	0.051	± 0.036	± 0.003
1957	73	0.097	± 0.045	± 0.005
1958	28	0.134	± 0.044	± 0.008

United Kingdom Samples[15]

	Mid and Late 1958		Jan - Jun 1959	
Age Group	No. of Samples	Mean Sr^{90} Conc. ($\mu\mu c$/g Ca)	No. of Samples	Mean Sr^{90} Conc. ($\mu\mu c$/g Ca)
Stillbirths	65	0.7	23	1.15
0-5 Years	27	1.5	21	2.8
5-20 Years	7	0.8	10	1.05
> 20 Years	7	0.10	3	0.08

in children, where growth was occurring in addition to exchange, higher concentrations were observed. Figure 9.2 shows the distribution curve of strontium-90 concentrations in bones of people in Western culture as a function of the age of the individual, calculated according to the considerations outlined in the discussion of diet concentrations[14]. The 1958 data on bone analyses were superimposed on the theoretical curves to illustrate the good agreement between the theoretical and practical results. The highest concentrations were in one year old children-about 2 μμc Sr^{90}/g Ca in 1958 compared with the concentration in adults of approximately 0.2 μμc Sr^{90}/g Ca. Adult average concentrations increased in 1959 to close to 0.3 μμc Sr^{90}/g Ca.

Future Strontium-90 Concentrations in Bone

It must be understood that the above type of distribution of strontium-90 concentrations in bone with age will not be preserved in the future. Rather, if the rate factor is as important as it is now surmised, one year old children in the future will not possess the maximum concentrations. Furthermore, the maximum concentrations of strontium-90 in the bones of these one year olds will reach a peak in 1961 and then rapidly decrease as the children become older. Actually in about 12 years time the concentrations in the bones of adults aged about 27 years will be higher than in the 13 year old youths at that time. These conclusions are illustrated in Figure 9.3. This curve shows the strontium-90 concentrations in people of Western culture in 1959 and the projected curves for various ages, based on the consideration that only 20 percent of the strontium-90 in the diet in 1959 was dependent on the cumulative deposit on the ground, and the remaining 80 percent

Fig. 9.2

CALCULATED CURVES OF EXPECTED STRONTIUM-90 CONCENTRATIONS IN BONES OF PEOPLE IN WESTERN CULTURE FOR 1957, 1958 AND 1959 (Taken from Kulp et al.[14])

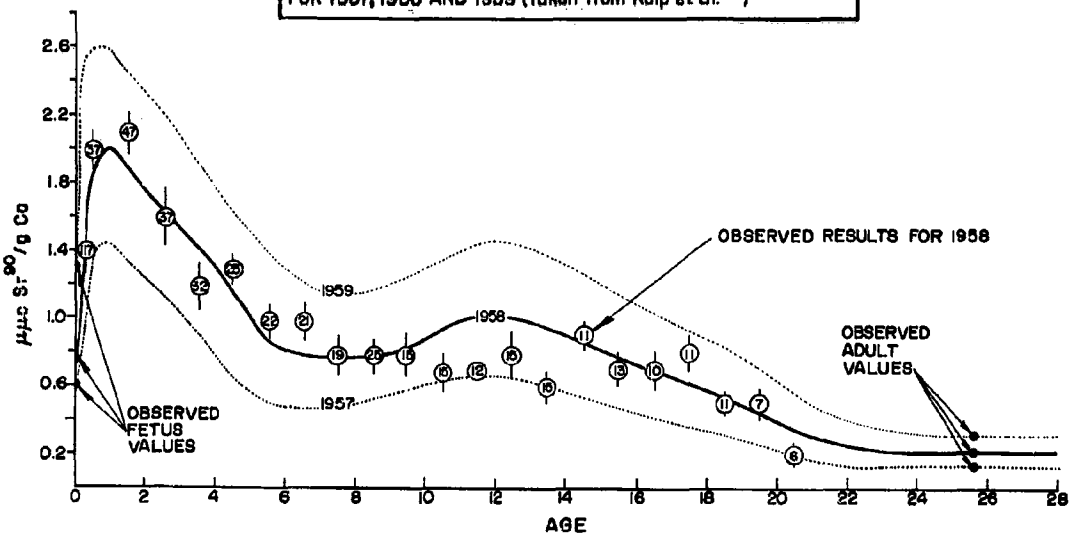
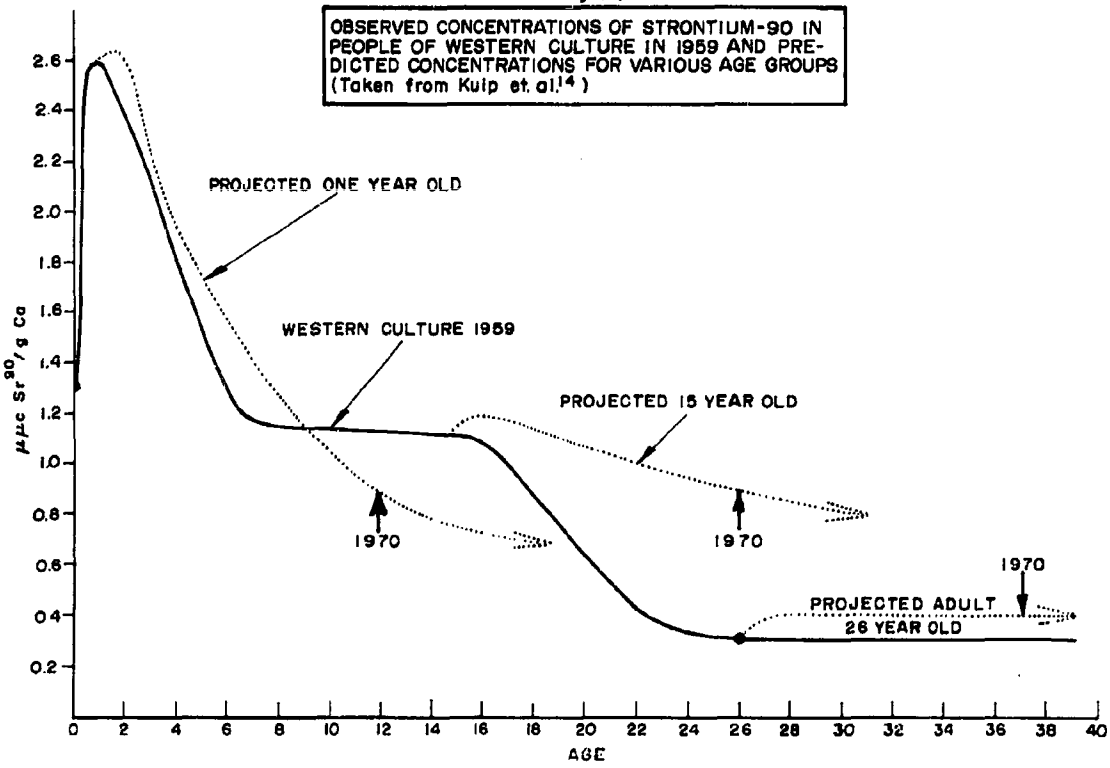


Fig. 9.3

OBSERVED CONCENTRATIONS OF STRONTIUM-90 IN PEOPLE OF WESTERN CULTURE IN 1959 AND PREDICTED CONCENTRATIONS FOR VARIOUS AGE GROUPS (Taken from Kulp et al.[14])



was derived from the fallout deposited during the previous twelve months[14]. The actual predicted concentration of strontium-90 in the diet under these conditions may be obtained from a plot similar to those shown in Figure 9. 1. These revised predictions result in lower concentrations of strontium-90 than had hitherto been calculated.

Summary of Strontium-90 Hazard

The hazard from strontium-90, both now and in future years, may be evaluated by comparing the concentrations in bone, which were described in previous sections, with the maximum permissible body burden of this nuclide as recommended by the National Committee on Radiation Protection[51]. Table 9. 6 summarizes the present position and what is believed to be the best estimates of future concentrations of strontium-90 in the body based on predictions shown in Figure 9. 3. With the exception of the present one year old children it is apparent that the majority of individuals have, or will have, less than 1 percent of the maximum permissible concentration of strontium-90 within their bones. The highest concentration of strontium-90 is in one year old children at the present time. Large differences in bone concentrations from areas with different diets are not expected even though the diet concentrations of strontium-90 in some areas may be several times higher than those in the United States. As an estimation of the variation in concentrations of strontium-90 in bone within a given population group in 1959, it was concluded by Kulp et al[14] that the standard deviation on the mean for adult subjects in Western culture was about 80 percent. All samples analyzed thus lay within a factor of 10 of the mean value, 97 percent within a factor of three of

Table 9.6 Present and Predicted Concentrations of Strontium-90 in Bones

	Sr^{90} Concentration μμc Sr^{90}/g Ca	%MPC
Industrial Maximum Permissible Concentration in Bone	2000	--
Large Population MPC in Bone	200	--
Sr^{90} Concentrations in Bone		
1. 1958 - Adults - World Average	0.20	0.10
1959 - Adults - World Average	0.28	0.14
1970 - Adults - Western Culture (30°N-50°N)	0.40	0.20
2. 1959 - 1 Year Olds - Western Culture	2.10	1.05
1970 - 12 Year Olds - Western Culture	0.9	0.45
3. 1959 - 15 Year Olds - Western Culture	1.1	0.55
1970 - 26 Year Olds - Western Culture	0.9	0.45

the mean, and 90 percent within a factor of two of the mean. Considerable fractions at the maximum permissible concentrations are therefore not expected on the basis of these results.

RADIATION HAZARDS FROM CESIUM-137

Cesium-137 (half-life ~30 years) is considered to be one of the most hazardous nuclides produced in the fission process. Together with strontium-90 this nuclide represents a major part of the total radiation hazard to man caused by nuclear explosions. Cesium-137 presents a hazard to man in two ways. Firstly, because of its chemical similarity to potassium, it is incorporated in soft tissue and can possibly lead to somatic effects. Secondly, it presents a genetic hazard because of its internal and external gamma irradiation of the gonads. The extent of the damage caused by chronic, low-level radiation exposure from both strontium-90 and cesium-137 is still unknown, but at least the concentrations of these nuclides both inside and outside the body are quite well established at the present time.

Similarly to strontium-90, the principal route of entry of cesium-137 into the body is through ingestion of foodstuffs. Only minor quantities of cesium-137 enter the body through drinking water and inhaled dust. Thus the concentrations of cesium-137 in body tissue will be dependent upon the concentrations of cesium-137 in the diet, the discrimination factors involved in going from diet to man, and the relative importance of the rate and cumulative factors governing the concentrations of cesium-137 in food.

Measurements of dietary concentrations of cesium-137 are few in number. Usually the concentrations of cesium-137 in milk and other dairy products are used to assess the dietary intake of this nuclide. Most of the following discussion of the cesium-137 contamination of foods is based therefore on the results of monitoring milk supplies.

Cesium-137 Concentrations in Milk

Extensive surveys of the cesium-137 concentrations in milk have been performed at several stations throughout the world. Table 9. 7 lists a number of these observations during the period 1956-1960. Because of the metabolic similarity of cesium and potassium the concentrations are conveniently expressed in $\mu\mu c\ Cs^{137}/g\ K$.

The most comprehensive studies of cesium-137 in milk and man have been performed by Langham and Anderson[16] at the Los Alamos Scientific Laboratory. Our discussion of the hazard from cesium-137 is confined to the conclusions of these workers since their findings are generally applicable to most of the data compiled at other laboratories.

It is clearly seen that there was a general rise in the concentration of cesium-137 in milk from 1956 until 1959. Results, which are available for the first six months of 1960, indicate that the average concentration of cesium-137 in milk dropped to approximately one half of the average concentration for 1959. During this latter period the rate of deposition of fallout also declined considerably (see Table 9. 4) while the total deposit of fallout on the earth's surface increased only slightly. Obviously, therefore, the concentration of cesium-137 in milk is

Table 9.7 Cesium - 137 Concentrations in Dried Milk (1956 - 1960)

Investigators	Country	Cesium - 137 Concentrations (μμc/g K)				
		1956	1957	1958	1959	1960
Langham and Anderson[16, 23, 24, 25]	U.S.A.	24	49*	57*	74*	38**
Booker[17]	U.K.		~ 10-140***	~10-100		
Anderson, Burton and Crookall[18, 19]	U.K.			~ 50-120	~ 30-160	
Mc Neill and Trojan[20]	Canada				~ 12-50	
Jensen[21]	Denmark	~ 20	~ 15-28	~ 5-33	~ 5-56	
Madshus and Baarli[22]	Norway			~ 10-55	~ 17-129	

* Weighted for population

** First six months - not weighted for population

*** Influenced by Windscale Accident - October 1957

largely dependent on the rate of deposition of cesium-137 on the grass by rainfall. The exact contributions of cesium-137 from the two sources, namely that which is deposited directly on the foliage and that which is derived through absorption from the soils cannot be calculated with these limited data. One reason why this is not possible is the fact that cesium-137 levels in milk in early 1960 were governed to a large extent by the amount of feed given to the animals in the winter of 1959-60, but which was grown in the summer of 1959. The preliminary data do suggest, however, that a maximum of 50 percent of the cesium-137 in milk in 1959 and 1960 was derived from the total cumulative deposit of cesium-137 in soils, the remainder being derived from recently deposited cesium-137. Continuous monitoring of the cesium-137 concentrations in milk during the years after 1959 will serve to clarify the respective contributions from these two sources.

Some consideration has already been given to future concentrations of strontium-90 in milk and diet, and Figure 9.1 shows the predicted values for the diet in $\mu\mu c\ Sr^{90}/g\ Ca$ for various contributions from the rate and cumulative factors. Cesium-137 concentrations in milk are expected to vary in a similar manner. Absolute values of the cesium-137 concentrations, expressed in $\mu\mu c\ Cs^{137}/g\ K$, are expected to be a factor of six higher than the strontium-90 concentrations, ($\mu\mu c\ Sr^{90}/g\ Ca$) as shown by current results[23] for this ratio in Table 9.8. Thus, the decrease observed in the preliminary results during 1960 for the cesium-137 concentrations in milk agree quite closely with results predicted on the assumption that 70 percent of the activity in milk in 1959 was derived from recent cesium-137 fallout. By 1964, if this trend be continued,

Table 9.8 Strontium-90 / Cesium-137 Ratios in Milk from Several Areas in the U. S. [23]

Area	Average Annual Rainfall (inches)	Sr^{90}/Cs^{137} Ratio $\left(\frac{\mu\mu c\ Sr^{90}/g\ Ca}{\mu\mu c\ Cs^{137}/g\ K}\right)$
Eastern U. S.	40 - 60	0.19 ± 0.02 *
Northwest Coast	40 - 60	0.14 ± 0.02
Midwest U. S.	20 - 40	0.19 ± 0.02
High Plains States	10 - 20	0.19 ± 0.03
West and Southwest	Irrigation	0.14 ± 0.03

* Standard error on mean

the average cesium-137 concentrations in milk from the 30°N-50°N latitude band will have dropped below 30 μμc Cs^{137}/g K.

Cesium-137 Concentrations in Humans

Several investigations of the cesium-137 concentrations in human beings resident in the Northern Hemisphere have been performed. The results of some of these analyses are shown in Table 9.9. Apparently the cesium-137 concentrations in man did not exhibit much change during 1956 and 1957, but subsequently they rose to a maximum in 1959 following the extensive nuclear testing which occurred in 1958. The data of Rundo[27] for body concentrations of cesium-137 in the United Kingdom during June 1960 show a significant drop from the results he obtained during December 1959-March 1960. This decrease was maintained during September 1960.

It is not justifiable to compare directly the data obtained on United States subjects with those obtained in the United Kingdom or elsewhere for a number of reasons. Firstly, the cesium-137 content of individuals is dependent upon the cesium-137 concentration in the diet and, since diets can vary in composition, it is expected that the body burdens of this nuclide can also vary. Secondly, the concentration of cesium-137 in the diet in a given area depends on the cumulative deposit and the rate of deposition of cesium-137 in the area, and because these latter factors are directly related to the annual distribution and quantities of precipitation, some variation in dietary concentration of cesium-137 is therefore expected. It is instructive, however, to estimate the contributions of milk and of the remainder of the diet to the body burden of cesium-137. The United States

Table 9.9 Average Cesium-137 Concentrations in Human Beings

Investigators	Country	Cesium-137 Concentrations ($\mu\mu c\ Cs^{137}/g K$)				
		1956	1957	1958	1959	1960
Langham and Anderson [16, 24, 25, 29]	U.S.	41	44	54	83	80
Maycock et al [26]	U.K.			~ 48	~ 44	
Rundo[27]	U.K.	~ 32	~ 37	~ 49	~ 59	~ 52
Onstead et al[28]	Germany				~ 80	

data used for this calculation are shown in Table 9. 10. Columns 2 and 3 list the average concentrations of cesium-137/g potassium in milk and humans in the United States for the period 1956-mid 1960. All concentrations were weighted according to population. The percent contribution of milk to the body burden of cesium-137 was calculated on the basis of 40 percent of total potassium intake by the body being derived from milk. A discrimination factor of 1. 8 in favor of cesium in going from milk to man was also assumed. Percentage contributions from milk to the total body burden of cesium-137 are shown in the final column. They vary from an estimated 34 percent for the preliminary data obtained in the first half of 1960 to 64 percent in 1959 and 80 percent in 1957. The concentrations of cesium-137 in the diet, excluding milk, shown in column 4, were calculated on the assumption that the remaining 60 percent of potassium in man was derived from this source and that the same discrimination factor of 1. 8 in going from diet to the body can be applied.

It is clear, if the data on cesium-137 in milk and the remainder of the diet are compared with the information on the rate of deposition of fallout as shown in Table 9. 4, that the concentrations of cesium-137 in milk closely follow the rate of deposition of this nuclide, while the concentrations in the remainder of the diet appear to lag behind the rate curve by about one year or slightly more. Further comparison of cesium-137 in the diet, excluding milk, and the average cumulative deposit of cesium-137 in soils in the 30°N-50°N latitude band (see Table 9. 4) during the period up to 1960 indicates that it is hardly possible to determine whether it is the rate or cumulative factor which primarily governs the

Table 9.10 Comparison of Cesium-137 Concentrations in Milk, Rest of Diet and in Humans in the U. S.

Year	Milk Concentration ($\mu\mu c\ Cs^{137}/g\ K$)	Concentration in Humans ($\mu\mu c\ Cs^{137}/g\ K$)	Calculated** Concentration in Rest of Diet ($\mu\mu c\ Cs^{137}/g\ K$)	Percent Contribution from Milk to Cs^{137} Body Burden
1956	24	41	21.9	42.2 %
1957	49	44	8.1	80.0 %
1958	57	54	12.0	36.0 %
1959	74	83	27.5	64.2 %
1960*	38	80	48.7	34.2 %
			Average =	51.3 %

* First part of the year

** 40% of dietary potassium intake from milk and a discrimination factor of 1.8 in favor of cesium in going from diet to man were assumed

concentration in this part of the diet. The weight of evidence from strontium-90 concentrations in the diet favors the former. Another year of observations, however, of cesium-137 in milk and humans or in total diet will serve to clarify this question.

Dose Rates from Cesium-137

(a) External radiation dose

The external genetic radiation dose rate from cesium-137 deposited in the 30°N-50°N latitude band may be calculated from the expression given by Langham and Anderson[16]:

$$*\text{Dose rate (mr/year)} = 5 \times 10^{-2}\, CE,$$

where C = surface deposit of cesium-137 (mc/mi^2)

and E = cesium-137 gamma ray energy (0.66 Mev).

Because of the limited data on cesium-137 in soils, an estimate of this quantity was made by multiplication of the strontium-90 deposits in this zone (Table 9.4) by the Cs^{137}/Sr^{90} ratio, which was assumed to be 1.8 ± 0.5. The resultant deposits and calculated dose rates are shown in Table 9.11. The infinite external dose from cesium-137 was calculated to be 138 mr for the mean life of cesium-137 ($\sim$43 years). Langham and Anderson[16] have assumed a conservative shielding factor (by buildings, dwellings, etc) of about 10, thus leading to an infinite external genetic dose of approximately 14 mr, or $\sim$ 0.5 percent of the genetic dose from natural background.

* United Nation's expression for the dose rate in mrem/year = $2 \times 10^{-2} C$.

Table 9.11. Estimated Average Cesium-137 Deposits in Soil in 30°N-50°N Latitude Band and the Resultant External Dose Rates

Date	Cs^{137} Deposit (mc/mi^2)	Dose Rate (mr/year)
1 July 1954	2.7	0.1
1 July 1955	11.0	0.4
1 July 1956	17.8	0.6
1 July 1957	28.1	0.9
1 July 1958	50.0	1.7
1 July 1959	78.5	2.6
1 July 1960	90.7	3.0
1 July 1961	92.3	3.0
1 July 1962	92.3	3.0
1 July 1963	91.6	3.0
1 July 1964	90.7	3.0
1 July 1965	89.3	2.9
1 July 1966	87.8	2.9
1 July 1967	86.2	2.8
1 July 1968	84.4	2.8
1 July 1969	82.8	2.7
1 July 1970	81.0	2.7

(b) Internal radiation dose

The International Commission on Radiological Protection has suggested that the internal radiation dose rate from cesium-137 may be calculated from the following expression:

$$q = \frac{2.8 \times 10^{-3}\ M\ W}{f_2 \sum E(\text{r. b. e.})N},$$

where q = total cesium-137 in the body (μc),

M = weight of critical organ (for cesium-137 this is the whole body i. e. ~ 70,000 g),

W = dose rate in rem/week,

f_2 = fraction of total body burden in critical organ,

$\sum E(\text{r. b. e.})N$ = average energy term veighted for the relative biological effectiveness and absorption in the critical tissue = 0.59.

Simplification of this expression for a 70 Kg man yields

$$*W = 2.2 \times 10^{-2} Q$$

where W is the dose rate in mrems/year and

Q is the cesium-137 concentration in the body in μμc Cs^{137}/g K.

A concentration of 50 μμc Cs^{137}/g K thus delivers a dose rate of about 1.1 mrems/year. Using the data given in Table 9.10 and assuming an average concentration of cesium-137 in humans of 20 μμcCs^{137}/g K for a two year period prior to 1956, it was calculated that the total dose delivered up to mid-1960 was approximately 7 mrems. Two extreme situations were then considered for the prediction of the infinite internal dose expected from cesium-137. In the first case it was assumed that the peak levels of cesium-137 attained in the body in 1959 and 1960

* Lindell's[31] expression, $1.5 \times 10^{-2} Q$, is somewhat less than this estimate.

were solely related to the cumulative deposit of cesium-137 on the ground. Thus the cesium-137 concentrations would decrease at a rate governed only by the half-life of the nuclide. A total dose of approximately 75-80 mrems was calculated. Together with the estimated 7 mrems dose during build up, the total internal dose from cesium-137 since the inception of weapons testing was estimated to be approximately 82-87 mrems.

If, in the other extreme situation, it is assumed that dietary concentrations, and hence body concentrations of cesium-137, are related only to the rate of deposition of the fallout, then an internal dose from cesium-137 of approximately 2 mrems is calculated for the period following mid-1960. For this second assumption, therefore, the total internal dose delivered by cesium-137 since the commencement of testing is about 9 mrems compared with the first estimate for the same quantity of just under 90 mrems.

Summary of Radiation Dosages from Cesium-137

The external and internal radiation hazards presented by cesium-137 are summarized in Table 9. 12. Here the present and predicted concentrations of cesium-137 in the body and on the surface of the earth are compared with the maximum permissible concentration in the body, as recommended by the NCRP[51], and the maximum permissible external dosage allowed to the most critical organs. As far as cesium-137 is concerned one of the most important considerations is the genetic dose. This dose is generally calculated for an exposure period of thirty years, and in the example chosen it was calculated for the most pessimistic situation, i. e. between 1959 and 1989 when surface concentrations of cesium-137 should

be their highest. It is obvious that the external genetic dose from cesium-137 over a thirty year period is a small percentage of the recommended maximum permissible dose for large populations, even when no attenuation of the gamma rays is assumed. Even if a conservative estimate that the attenuation is a factor of ten be hypothesized, the percentage of the maximum permissible dose is only 0.04. The internal concentrations of cesium-137 in the body are likewise quite small fractions of the permissible levels. Indeed, if the rate of deposition of fallout is the most important factor governing body concentrations of cesium-137, a rapid decrease in the percentages of permissible concentrations, as shown in Table 9.12, will occur within the next few years and will approach zero by about 1965, unless weapon testing is resumed.

RADIATION HAZARD FROM CARBON-14

Naturally-occurring carbon, an element which is essential to all living matter, is comprised of three isotopes: carbon-12, a stable nuclide, carbon-13, also a stable nuclide whose abundance is about one percent of the former, and lastly carbon-14 a radioactive nuclide which occurs approximately one in 10^{12} carbon-12 atoms. Carbon-14 decays by β^- emission according to the following scheme:

$$C^{14} \longrightarrow N^{14} + \beta^- + 0.155 \text{ Mev}.$$

The mean energy of β^- decay is 0.050 Mev, as compared to the maximum energy 0.155 Mev recorded in this equation. In addition to the possible dangers resulting from the ionizing radiation produced in the above reaction, hazards may arise from each or all of the following:

Table 9.12 Present and Predicted Radiation Hazard from Cesium-137

	(a) Internal Concentrations of Cesium-137	$\mu\mu c\ Cs^{137}/g\ K$	%MPC
	Industrial Maximum Permissible Concentration of Cesium-137 in Whole Body *	2.1×10^5	
	Large Population MPC in Whole Body	2.1×10^4	
	Observed Concentration in Whole Body (1959)	$\sim$ 80	0.4
**	Predicted Concentration in Whole Body (1989)	$\sim$ 40	0.2
***	Predicted Concentration in Whole Body (1989)	0	0

	(b) External Radiation Dose	mrems	%MPC
	Industrial Maximum Permissible Dose from Cesium-137 (over 30 years)	150×10^3	
	Large Population MPD from Cesium-137 (over 30 years)	15×10^3	
	Average External Genetic Dose (1959-1989) from Cesium-137 in 30°N - 50°N Latitude Band (no attenuation assumed)	$\sim$ 60	0.4

* Standard Man of 70kg and 140g Potassium assumed.

** Calculated assuming cumulative deposit of Cs^{137} governs body concentration of Cs^{137}

*** Calculated assuming rate of deposition of Cs^{137} governs body concentration of Cs^{137}

a. the chemical change from carbon-14 to nitrogen-14 which occurs during the β^- decay,

b. the recoil energy of the nucleus after emission of the β^- particle,

c. the residual electronic energy within the atom after the release of the β^- particle.

The half life of carbon-14 is $\sim$5600 years. Because of this rather long life, compared with that of other nuclides, such as strontium-90 and cesium-137 ($t_{1/2} \sim$ 28 years), one of the most important considerations to be borne in mind is the total genetic dose delivered by carbon-14 To compute this dose, however, it is necessary to know the carbon-14 concentrations within the biosphere at the present time and be able to predict future concentrations accurately.

As a result of nuclear weapons testing the total inventory of carbon-14 has increased substantially, and since the early 1950's this additional carbon-14 has been sufficient to cause measurable increases above the natural concentrations already present within the atmosphere and biosphere[32, 33, 34]. This radiocarbon from nuclear tests arose, not as a direct product of the fission process, but from the side reaction of neutrons, produced during the detonations, with the nitrogen of the atmosphere. Bomb-produced radiocarbon will not, of course, be distributed immediately within the exchange reservoirs in the same manner as naturally occurring carbon-14. Rather, because of the finite times involved in the exchange processes which occur between these reservoirs, such as for example between the atmosphere and the oceans, transient conditions will exist for several years. Fortunately it is possible to estimate what these transient conditions will be from our knowledge of the behavior of naturally occurring carbon-14. From our information on these conditions and estimates of the total

amount of carbon-14 produced in weapons testing to date it is possible to make some assessment of the hazard from bomb-produced carbon-14.

Distribution of Natural Carbon-14 Within the Carbon Cycle

The distribution of natural carbon-14 within the several possible exchangeable carbon reservoirs is shown in Table 9.13. Carbon within the sedimentary carbon reservoirs is not considered in this discussion because the time scale involved for exchange between the atmosphere and the sedimentary reservoir is much larger (millions of years) than the exchange times involved in the reservoirs shown in Table 9.13. Within the exchange reservoirs, however, it is clear from this table, which depicts the final equilibrium situation, that most of the carbon-14 is in the ocean. This is the situation which existed immediately before 1900 and is believed to have existed in this way for many thousands of years prior to this date. Since 1900, however, the steady state situation has been disturbed in two ways. Firstly, considerable quantities of carbon-14 free carbonaceous materials from the sedimentary reservoir have been oxidized to carbon dioxide, and this reduced the carbon-14 concentration in the reservoirs, which have exchange constants comparable to the time period over which this process has been going on, i.e., over the past 60-70 years. Secondly, the testing of nuclear weapons has added large quantities of carbon-14 to these same reservoirs and increased the concentration of this nuclide. These three periods, in which we have experienced

a. steady state distribution of C^{14},

b. decreases in C^{14} concentrations (Suess Effect[35]) and

Table 9.13 Distribution of Naturally-Produced Carbon-14 within Exchangeable Carbon Reservoirs

Carbon Reservoir	Mass of Carbon g/cm^2 Earth's Surface	Total Number of C^{14} Atoms in Reservoir (x 10^{-27})
Atmosphere	0.125	36
Biosphere	0.06	18
Humus	0.20	50
Surface Ocean Water	~ 0.18	50
Remainder of Ocean	~ 7.50	1950
Totals	8.06	2104

c. increases in C^{14} concentrations as a result of nuclear weapons testing, are depicted in Figure 9. 4.

It is the hazards resulting from the increased carbon-14 concentrations experienced from the early nineteen fifties up to the present time with which we are mainly concerned in this section.

Present and Future Concentrations of Carbon-14 in the Atmosphere

The present distribution and the total amount of carbon-14 of nuclear bomb origin is fairly well-known at the present time. Figure 9. 4 indicates the increase in carbon-14 concentration in the troposphere of the Northern Hemisphere up to 1959 when the concentration was on the average about 28 percent higher than pre-1900 wood. In the Southern Hemisphere the carbon-14 concentrations had attained a value about 18 percent higher than pre-1900 wood. The difference between the hemispheres arose because of the preponderance of nuclear testing conducted in the Northern Hemisphere. Even within the Northern Hemisphere there is definite evidence for a latitudinal variation in carbon-14 concentrations. For example, in 1959 a range of carbon-14 concentrations from 26 percent above pre-1900 wood concentration at $41^{o}N$[36] to 33. 3 percent at $56^{o}N$[37] was observed. This variation was attributed to the rapid deposition in 1959 of nuclear debris from Soviet tests conducted in the Arctic regions in the fall of 1958. Such large variations, within and between the hemispheres, should have decreased considerably by 1961 and are not expected to occur again in the future, primarily because of the fairly rapid mixing of air across the equator and the continued moratorium on nuclear testing.

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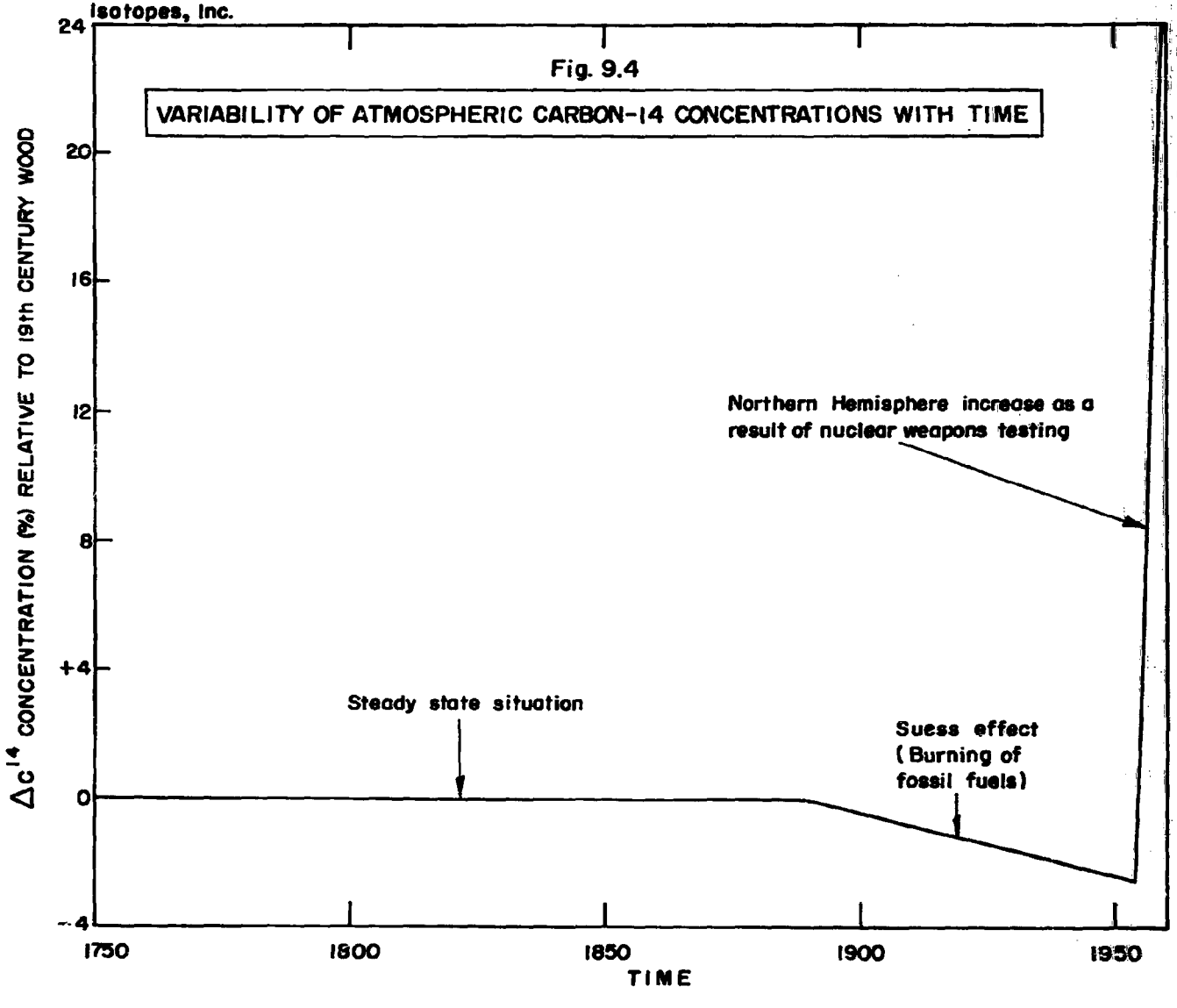
Fig. 9.4

VARIABILITY OF ATMOSPHERIC CARBON-14 CONCENTRATIONS WITH TIME



The concentrations and the total inventory of carbon-14 in the stratosphere have also been measured. Estimates of the stratospheric inventory of carbon-14 during the period 1 July 1955-1 July 1958 yielded results ranging from 5.6×10^{27} to 8.6×10^{27} carbon-14 atoms. Following the extensive testing in 1958, it was estimated that on July 1, 1959 between 10 and 13×10^{27} carbon-14 atoms remained in the stratosphere[38]. At the same time it was calculated that the troposphere contained 6.7×10^{27} carbon-14 atoms (equivalent to an average world-wide increase of ~22 percent over the pre-1900 concentration), the oceans 2.2×10^{27} carbon-14 atoms and the biosphere 0.4×10^{27} carbon-14 atoms, giving a total of between 19 and 22×10^{27} carbon-14 atoms produced to date (Table 9.14). There is some question as to the accuracy of the total inventory of carbon-14 produced to date by nuclear testing but this will not be discussed here. At the present time it appears that the best estimate which can be made leads to a value of 21×10^{27} carbon-14 atoms, and this is used in subsequent evaluations of the dose. Before proceeding to this question, however, it is necessary to evaluate how the distribution of carbon-14, as given in Table 9.14, will change with time. Obviously the variation of the carbon-14 concentrations within the troposphere, and thus within the body, is the important factor, since the time lag for the transfer of carbon from air to the blood stream is short (~one year).

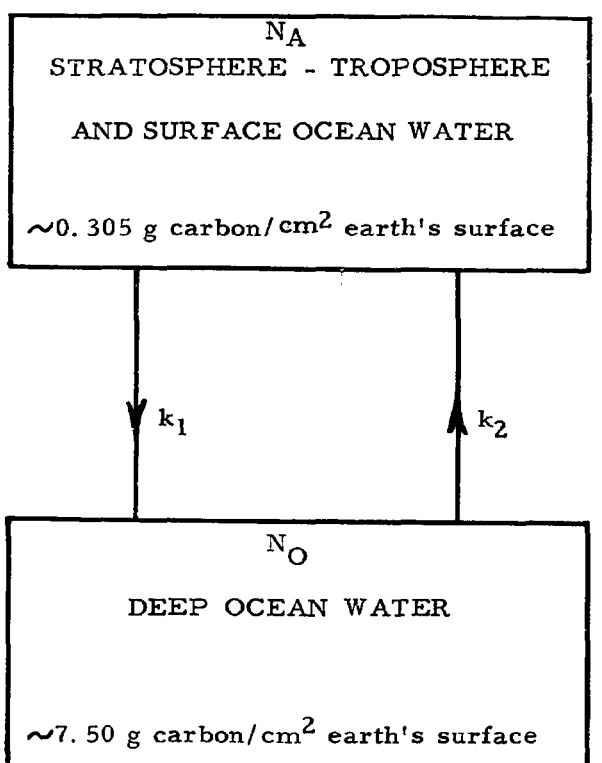
Various models have been used in the past to describe the future concentrations of bomb radiocarbon within the numerous exchange reservoirs. It appears, however, that a simple system involving two reservoirs (Figure 9.5), namely the atmosphere-surface ocean water and the deep oceans is adequate for

Table 9.14 Estimated Distribution of Bomb-Produced Radiocarbon within the Various Exchange Reservoirs on July 1, 1959

Reservoir	Carbon-14 content (Atoms x 10^{-27})
Stratosphere	10 - 13 *
Troposphere	6.7
Biosphere	0.4 *
Ocean	2.2 *
Total	19 - 22

* Calculated Values

Figure 9.5. Reservoir Model for Prediction of Future Concentrations of Carbon-14 from Nuclear Testing



the purpose of making fairly accurate estimates of the dosages from weapons-produced carbon-14. In this simple model the stratosphere, troposphere, and surface ocean water are considered to be in rapid exchange with each other and can therefore be considered as a single reservoir. The second reservoir is taken to be the deep ocean water which exchanges at an extremely slow rate with the atmosphere-surface ocean system. Measurements in the pre-bomb era suggest that the age of deep ocean water is, on the average, about 1200 years. Using the notations shown in Figure 9.5, k_2 must therefore be 0.00083 year $^{-1}$ and k_1 = 0.020 year $^{-1}$. The rate of change of the bomb-produced carbon-14 (N_A) in the atmosphere ocean water system may then be written as

$$\frac{d N_A}{dt} = -k_1 N_A + k_2 N_O - \lambda N_A \qquad (1)$$

where λ is the decay constant.

$N_A + N_O = N_T e^{-\lambda t}$, where N_T = total amount of bomb-produced radiocarbon at t = 0. This equation may be solved for N_A, yielding

$$N_A = \frac{N_T}{k_1+k_2} \left\{ k_1 e^{-(k_1 + k_2 + \lambda)t} + k_2 e^{-\lambda t} \right\} , \qquad (2a)$$

or

$$N_A = N_T \left\{ \frac{k_1}{k_1 + k_2} e^{-(k_1 + k_2 + \lambda)t} + \frac{k_2}{k_1+k_2} e^{-\lambda t} \right\}. \qquad (2b)$$

Substituting values for k_1, k_2 and λ

$$N_A = N_T \left\{ 0.96\, e^{-0.021t} + 0.04 e^{-0.00012t} \right\} . \qquad (3)$$

Thus the change in carbon-14 concentrations within the atmosphere can be considered for two fractions, given by the two terms in this equation. One of these represents the changes in concentration as equilibrium with the deep ocean water is approached, and the second term represents the radioactive decay of carbon-14 within the overall system after equilibrium is attained. Equation (3) is plotted in Figure 9.6 where N_A is given in percent difference from the pre-1900 carbon-14 concentration in the atmosphere. An initial value for N_T of 21×10^{27} carbon-14 atoms has been assumed. For the purpose of this discussion this bomb-radiocarbon may be considered as having been injected as a single pulse in 1958.

Dosages from Bomb-produced Radiocarbon

Calculations of the dosages from carbon-14 can be made from a modification of equation (3) by relating N_T to the dose rate from natural radiocarbon. It can be shown that the dose rate in the body from natural radiocarbon, which has a specific activity of 14 dpm/g carbon, is approximately 1.1 mrads/year. Since N_T is approximately 24 percent above the natural level of carbon-14 activity, the dose rate from bomb-produced radiocarbon at any time in the future is given by

$$D_R = 0.24 \left(0.96\, e^{-0.021t} + 0.04\, e^{-0.00012t}\right),$$

and the total dose over a time period T (years) from 1958 is given by

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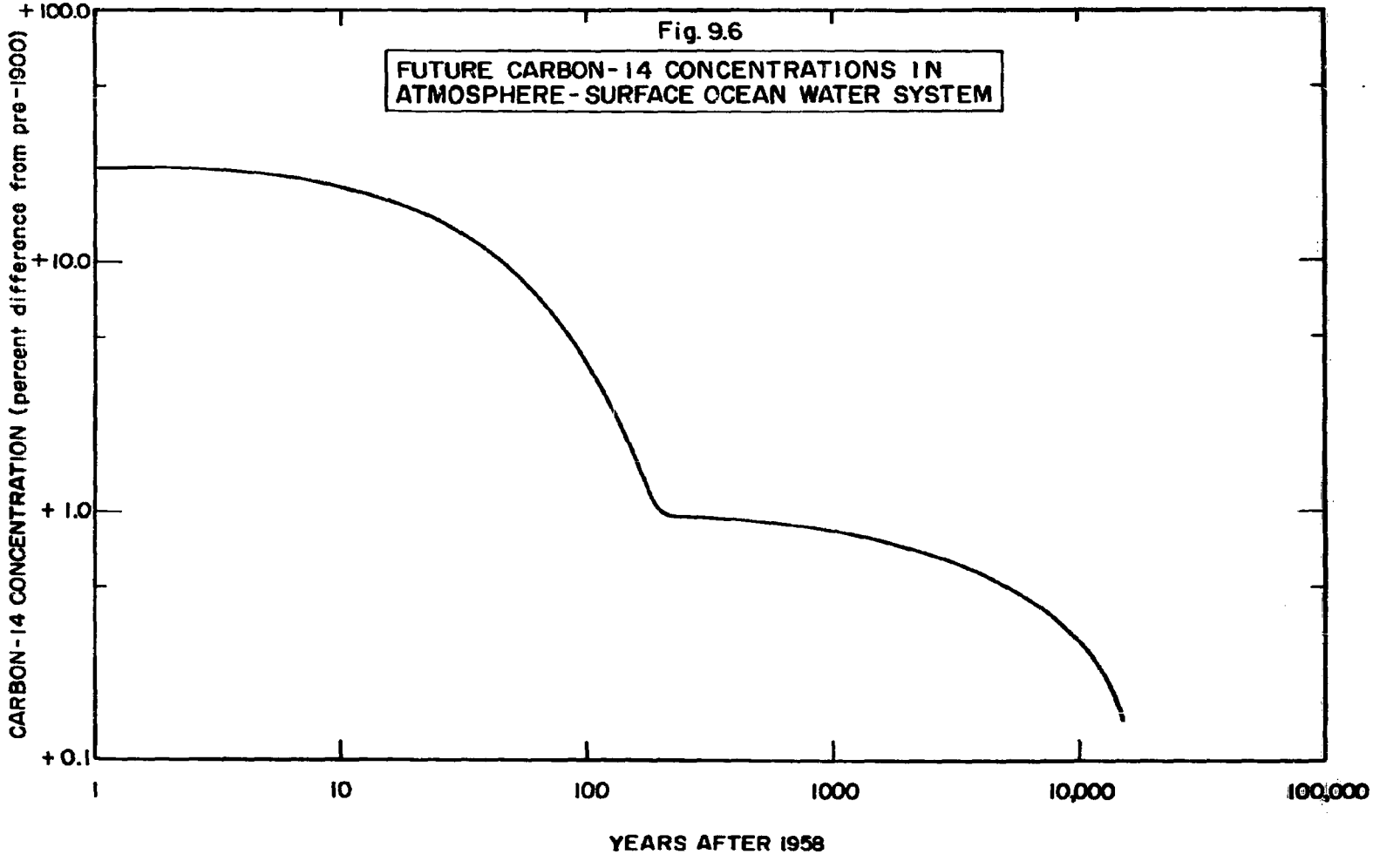
Fig. 9.6

FUTURE CARBON-14 CONCENTRATIONS IN ATMOSPHERE-SURFACE OCEAN WATER SYSTEM



$$D = 0.24\left\{\frac{0.96(1-e^{-0.021T})}{0.021} + \frac{0.04(1-e^{-0.00012T})}{0.00012}\right\}$$

$$\approx 11\ (1-e^{-0.021T}) + 80(1-e^{-0.00012T}) . \qquad (4)$$

The thirty year dose to the body from carbon-14 is, therefore, about 6 mrems and the infinite time dose is about 90 mrems. Direct comparison of these results with similar data on cesium-137 may be made. A total internal dose from this nuclide was calculated to lie between 9 and 90 mrems depending upon whether the dietary concentrations of cesium-137 were governed by either the rate of fallout or the cumulative deposit of fallout, respectively. In addition the maximum thirty year external dose from cesium-137 was calculated to be 6 mrems if an attenuation factor of ten was assumed. The infinite external dose from cesium-137 will not be much greater than 6 mrems because the mean-life of this nuclide is about 43 years. Obviously, therefore, carbon-14 presents a hazard which is certainly as important as cesium-137 from the genetic standpoint, even if the cumulative factor plays an important part in controlling the intake of cesium-137. On the other hand, carbon-14 assumes a more important role than cesium-137 and all other fission products combined if the rate factor governs the intake of radioactivity in food products and the infinite dose from carbon-14 delivered to many generations is the dose of consequence.

RADIATION HAZARD FROM PLUTONIUM

Early studies of the passage of plutonium through the ecological cycle from soils to man revealed rather large discrimination factors against the transfer of plutonium to the human body. Thus the ingestion of plutonium, calculated from theoretical estimates of the amount of fallout plutonium on the earth's surface, was not assessed as being a major hazard. The cause of concern for plutonium, however, arose originally as a result of inhalation studies by Stannard[39] and Bair[40], which indicated accumulation of this activity in the pulmonary lymph nodes. Stannard[39], in particular, concluded from his work that future calculations of the maximum permissible doses should be performed on the basis of the pulmonary lymph nodes being considered as the critical organ. Further investigations of inhalation as the mode of entry of fallout plutonium into man was prompted by estimates of the concentration of plutonium in ground level air. It was calculated that in 1957 the plutonium concentration in ground level air in Washington, D. C. was $\sim$15 dpm per 100 standard cubic meters, or approximately 0.2 percent of the maximum permissible concentration[41]. In 1959 samples from the same area were approximately 1 percent of the maximum permissible concentration. These data led us to a preliminary investigation of plutonium in human and animal tissue in 1959*[42]. Unfortunately, however, virtually no additional results have been reported in the literature during the past year, although some attempt was made to document fallout levels of plutonium in precipitation samples.

* A more detailed account of this work appears in Part III of this report.

The results of plutonium analyses of human tissue from residents of the New York City area are shown in Table 9.15. It is clear from the data on composite organs that the lung contains by far the majority of the plutonium activity, thus giving some support to the theory that inhalation is the significant mode of entry of fallout plutonium into man. Because of the relative sizes of the various organs, however, it appears that the highest concentrations of plutonium occur in the lymph system and the gonads. Consequently the highest percentages of the maximum permissible doses are delivered to the gonads and the lymph node system. Comparisons may be made with the other probable hazardous products of nuclear detonations. For example, in the case of strontium-90 (see previous sections), it was stated that in 1959 the strontium-90 concentrations in the bones of adults living in the Northern Hemisphere was about 0.30 $\mu\mu$c Sr^{90}/g Ca, or 0.15 percent of the maximum permissible concentration of this nuclide in the skeleton. Thus the percentage of the maximum permissible dose delivered by plutonium to the lungs, lymph nodes, and gonads ranges from 1/3 to 10 times the fraction of the maximum permissible dose delivered by strontium-90 to the skeleton.

If, as suggested above, inhalation is the important method of incorporation of plutonium in man, it would be expected that concentrations of plutonium in the various organs of the body would decrease as the rate of fallout of plutonium decreased. In Chapter 7 we have shown that during 1960 the average rate of deposition of strontium-90 decreased considerably from the rate experienced in 1959. Furthermore, it is clear that concentrations of

Table 9.15 Plutonium Analyses of Human Tissue**

Sample*	Year of Death	Sample Weight (g)	DPM ± Stnd. Dev.	DPM/g of Tissue ± Stnd. Dev. (10^{-3})	%MPD ± Stnd. Dev.
lung - 1	1953	950	0.409 ± 0.030	0.430 ± 0.037	0.012 ± 0.001
lung - 2	1954	1000	0.760 ± 0.062	0.760 ± 0.062	0.022 ± 0.002
lung - 3	1958	398	4.75 ± 0.44	11.9 ± 1.1	0.34 ± 0.03
tracheo-hilar lymph nodes-3	1958	3.4	0.22 ± 0.11	65 ±32	1.9 ± 0.9
lung -4A	1959	450	0.565 ± .007	1.26 ± 0.17	0.036 ± 0.005
4B	1959	450	0.611 ± .077	1.36 ± 0.17	0.039 ± 0.005
Composite Organs - Group 1					
lung	1959	2340	4.06 ± 0.39	1.74 ± 0.17	0.050 ± 0.005
pulmonary lymph nodes	1959	53.6	0.57 ± 0.21	11 ±4	0.32 ± 0.11
spleen	1959	889	1.02 ± 0.19	1.15 ± 0.21	0.033 ± 0.006
kidney	1959	949	0.94 ± 0.19	0.99 ± 0.22	0.028 ± 0.006
gonad	1959	96	0.76 ± 0.18	7.9 ± 1.9	0.68 ±0.16

* Samples collected from 1953 to 1958 were made available by Dr. Arthur Schulert, Lamont Geological Observatory, Palisades, New York
Samples collected in 1959 were supplied by Dr. Roth, Head Pathologist, Bergen Pines Hospital, Oradell, N. J.

** Performed at Isotopes Inc., Westwood, N. J.

plutonium in air and precipitation are rapidly approaching the immeasurable stage with present experimental techniques. Provided that there is little resuspension of plutonium from soils, etc. and that the Pu/Sr^{90} ratio in fallout has not changed considerably during the past year or so, it is indeed questionable whether any experimental program designed to clarify some of the problems of present fallout plutonium deposition and its entry into man would be fruitful. What does remain to be done, however, is a comprehensive investigation of the concentrations of plutonium in man resulting from past weapons testing and studies of the critical deposition sites and possible translocations within the body.

RADIATION HAZARD FROM OTHER SOURCES

In addition to the radiation of man from the long-lived products of nuclear weapons testing, there are other contributions to the total dose which stem primarily from the shorter-lived isotopes. In the past these nuclides were considered to be important only in tropospheric fallout because of the relatively long residence times assumed for stratospheric fallout. With the downward revision of these residence times[1], however, attention was focused once again on the possible hazard presented by the short-lived components of fallout[53].

The radiation dose from the short-lived nuclides may be divided into two fractions according to the radiation source, i. e., the external or internal source, and the latter may be further subdivided according to the mode of entry of the radioactivity into humans, i. e., via inhalation or ingestion.

External Radiation from Short-lived Products of Nuclear Detonations

The United Nations[30] reported that the level of short-lived activity deposited on the ground in the Northern Hemisphere was maintained at approximately 50 to 200 mc/km^2 (or 130 to 520 mc/mi^2) up to the end of 1957. This activity concentration corresponds to a dose rate to the gonads of about 0.25 to 1 mrem/year if an average gamma ray energy of 0.5 Mev and an attenuation factor of 10 are assumed. During the period of nuclear testing up to the end of 1957, therefore, the total dose to the gonads was between 1 and 5 mrems. Considerable nuclear testing occurred in 1958, and the fission yield during this latter period was a large fraction (about one-third) of the yield up to the end of 1957. Furthermore, data on the presence of fission products in precipitation in 1959 indicate that the material from Soviet tests performed in 1958 was rapidly deposited in the Northern Hemisphere[43]. Thus the radiation dose from tests conducted in 1958 would probably be close to the dose from previous tests. Hence, it is estimated that the average external dose from short-lived nuclides in the Northern Hemisphere is probably less than 10 mrems. For comparison it may be noted that average cesium-137 concentrations in the 30°N-50°N latitude band led to a calculated external dose of 6 mrems for the thirty year period between 1959 and 1989.

Internal Radiation from Inhalation of Short-lived Products of Nuclear Detonations

The internal radiation from short-lived nuclides is derived from radioactivity entering the body by two modes, inhalation and ingestion. Inhalation radiation dosages to the body have been computed by Marley and Pochin[44] for an

average concentration of 10^{-15} curies of fission product per liter of air, which was an average activity level experienced in the Northern Hemisphere during 1956-1957. The results of the calculations of dose rates, which are shown in Table 9.16, were computed using I. C. R. P. criteria for retention, volume of inhaled air, weight of critical organs, etc. During 1958 and 1959 the total beta activity of ground level air in the vicinity of Washington, D. C. showed an average concentration which was about three times higher than in 1956-1957. Thus, if it is assumed that the concentrations observed in 1956-1957 were maintained during 1954-1957 and were tripled during 1958 and 1959, the total dose (in mrems) delivered to the various organs may be obtained by multiplication of the dose rates shown in Table 9.16 by a factor of ten. Maximum permissible dose rates for the general population are given by the N. C. R. P. [51] as 1.5 rem/year for most individual organs of the body, 3.0 rem/year when the critical organ is the thyroid or skin, and 0.5 rem/year when the gonads or the whole body is the critical organ. In 1958 and 1959, therefore, the average dose rate to the lungs, if the radioactive material is insoluble, was 0.3 percent of the maximum permissible dose rate. Even during the spring of 1959, when concentrations observed in ground-level air at Miami reached 20-25 x 10^{-15} curies/liter, the temporary dose rate to the lungs, if the material was considered insoluble, was 2-2.5 percent of the maximum permissible dose rate.

Of particular interest in the discussion of the inhalation of short-lived nuclides is iodine-131. It is generally accepted that inhalation is the main portal of entry of iodine-131 into man, and the interest in this nuclide stems primarily

Table 9.16 Annual Dose Rates from Inhaled Fission Products During 1956-1957. (Air Concentration = 10^{-15} curies/liter)

Organ	Dose Rate (mrem/year)
Whole Body	0.2
Lung (if radioactive material is soluble)	0.1
Lung (if radioactive material is insoluble)	1.5
Thyroid	0.6
Bone	0.15
Average Bone Marrow	0.05
Average Gut	0.03

from its selective concentration in the thyroid glands. Considerable work on iodine-131 concentrations in both cattle and human thyroids has been performed since 1954. During 1955 and 1956[45] the average concentration in humans was approximately 2 μμc per gram of thyroid, and in later periods of rather intensive testing e. g., in December 1958-January 1959 concentrations rose as high as 14-23 μμc per gram of adult thyroid tissue[46, 47]. The adult human thyroid weighs approximately 20 grams and the maximum deposit permitted in the thyroid is 7×10^4 μμc. Thus the levels of iodine-131 in human thyroid in 1958-1959 were a small fraction (0. 4-0. 7 percent) of the maximum permissible concentration assigned for this nuclide. Higher concentrations, up to 265 μμc per gram of thyroid tissue, were observed in human fetuses in May 1959 when fallout rates were at their highest. From these observations on fetuses, Beierwaltes et al[47] calculated that the maximum total radiation dose delivered to the fetal thyroid gland was 0. 47 rad and the average dose was 0. 05 rad . If a RBE for iodine-131 radiation of one is assumed, these results correspond to effective dosages of 0. 47 rems and 0. 05 rems to the thyroid, which may be compared to the I. C. R. P.'s upper limit for a 13 week dose to the thyroid of 0. 8 rems.

Generally speaking, therefore, the dose rates and dosages from inhaled short-lived nuclides to various parts of the body, when considered for the period from the commencement of nuclear testing to the present time, were small fractions of the maximum quantities permitted. When these factors were considered over shorter periods, however, considerable fractions of the maximum permissible concentrations were attained. Considerable attention ought to be given, therefore, to the possible hazards from short-lived nuclides during prolonged

periods of nuclear testing and, of course, in the event of nuclear war.

Internal Radiation from Ingestion of Short-lived Products of Nuclear Detonations

The contribution to the total dose to the human body from the ingestion of short-lived nuclides is extremely uncertain. There are very few data on their concentrations in foodstuffs, only isolated results being available on zinc-65[48, 59], barium-140[50], and strontium-89 contents of individual products[2, 10]. Zinc-65 ($t_{1/2}$ = 245 days) is, of course, somewhat longer-lived than the other two and has been detected in foods and people from the environment of the Hanford reactor site[48]. More recent investigations[49] revealed the presence of zinc-65 in a wide variety of foods grown in areas which are believed to be uninfluenced by reactor effluents. The concentrations in these latter materials were naturally much lower than those collected near Hanford. Maximum concentrations in foods, reported for these more normal conditions, were approximately 0.003 percent of the maximum permissible concentrations. Peak barium-140-lanthanum-140 activities observed in milk in 1957 were about 2 mμc per liter of whole milk, which is approximately 7 percent of the maximum permissible concentration of 30 mμc per liter. Subsequent barium-140-lanthanum-140 concentrations probably increased above those concentrations observed in 1957, but it is unlikely that they were more than three or four times higher. Other contributors to the diet will probably have much lower concentrations of barium-140 because of their age, which is old compared to the half-life of barium-140 (12.4 days).

Again it appears that the concentrations of radioactivity from relatively short-lived products of nuclear detonations are quite small fractions of maximum

permissible concentrations. The detection in foodstuffs of activities with half lives less than about 100 days is quite difficult because of the small activities involved and the fact that the average age of the diet is such that the activities will have decayed considerably before measurement. Even though high percentages of the maximum permissible concentrations of short-lived products were observed in certain areas in the past they were maintained only for short periods and have now dropped to immeasurable levels.

RADIOACTIVITY AND THE BIOLOGICAL HAZARD

In previous sections the concentrations in the environment of several radioactive products of nuclear detonations were examined. Table 9.17 summarizes these concentrations as fractions of the maximum permissible concentrations, which have been suggested by the N. C. R. P. as applicable to the general population[51]. In relative terms it is apparent that strontium-90 is potentially the most hazardous nuclide, giving rise to the highest sustained fractions of the maximum permissible concentration. During the period of weapons testing the short-lived nuclides produced relatively high concentrations both inside and outside the human body, but, following the cessation of atmospheric testing in 1958 and the spring deposition of fallout in 1959, these contributions to the total dose became negligible. From the genetic standpoint there is some question concerning the most hazardous nuclide. A total genetic dose delivered by carbon-14 over its lifetime was shown to be in excess of that delivered by all other products of nuclear testing. The question remains, however, whether the dose from carbon-14, which will be

Table 9.17 Summary of Average Concentrations of Radioactivity in Man from Fallout

Nuclide	Period	Conditions	Percent of MPC
Strontium-90	1959	Adults - World Average	0.14
	1970*	Adults - Western Culture	0.20
	1959	1 Year Olds - Western Culture	1.05
	1970*	12 Year Olds - Western Culture	0.45
	1959	15 Year Olds - Western Culture	0.55
	1970*	26 Year Olds - Western Culture	0.45
Cesium-137	1959 - 1989	External Dose to Gonads (30°N-50°N) Shielding Factor = 10	0.04
	1959	Internal Dose to Whole Body (30°N-50°N)	0.40
	1989	Internal Dose to Whole Body (30°N-50°N) Concentrations Governed by Cumulative Deposit of Cs^{137}	0.20
	1989	Internal Dose to Whole Body Concentrations Governed by Rate of Deposition of Cs^{137}	0
Carbon-14	1959	Northern Hemisphere	0.10
	2000	World Troposphere - allowing for Burning of Fossil Fuel	0
Plutonium	1959	Scattered Samples New York Area	
		(1) Lungs	0.05
		(2) Pulmonary Lymph Nodes	0.32
		(3) Gonads	0.68
	1989		?
Other Nuclides	Up to 1958	Average External γ Dose	0.05 - 0.20
Primarily Short-Lived Products e.g. Ba^{140}, Sr^{89}, Zr^{95}-Nb^{95}, Ce^{144}, Ru^{106}, etc.	1958-1959	Average Externalγ Dose	0.2 -1.0
	1956-1957	Average Internal Inhalation Dose	~ 0.10
	1958-1959	to Lung - Material Insoluble	~ 0.30
	Spring 1959		2-3
Iodine-131	1955-1956	To Thyroid - Adults	0.003
	1959	To Thyroid- Adult Maximum	0.4 - 0.7

* Future concentrations calculated on the assumption that in 1958 20% of the Strontium-90 in the diet resulted from the cumulative deposit of Strontium-90 in the soils.

spread over 250-300 generations, can be directly compared with other dosages delivered over only 2 or 3 generations[38]. In addition the dosage from plutonium remains to be defined more accurately.

A complete discussion of the biological consequences of these concentrations of radioactivity is beyond the scope of the present discussion. However, a few pertinent remarks and a very brief outline of current theories on the effects of atomic radiations are given for completeness.

It is still apparent that although there is a considerable wealth of information concerning the effects of large doses of radiation on animals, and in some cases on humans, our knowledge of the effects of chronic irradiation of humans or animals at low activity levels is practically nil. According to one theory any dose of ionizing radiation to the human body, no matter how small, will produce some effects. On the other hand, a second theory maintains that a threshold dosage exists below which somatic damage will not manifest itself during the life of an individual (see Figure 9.7). Proponents of this theory, however, generally agree that genetic effects will occur with even the smallest dosages.

It is obvious that one cannot wait for many generations of humans to ascertain the genetic effects of fallout radiation. Scientists have resorted, therefore, to experiments involving animals which can produce many generations in a relatively short time. Unfortunately these experiments always present

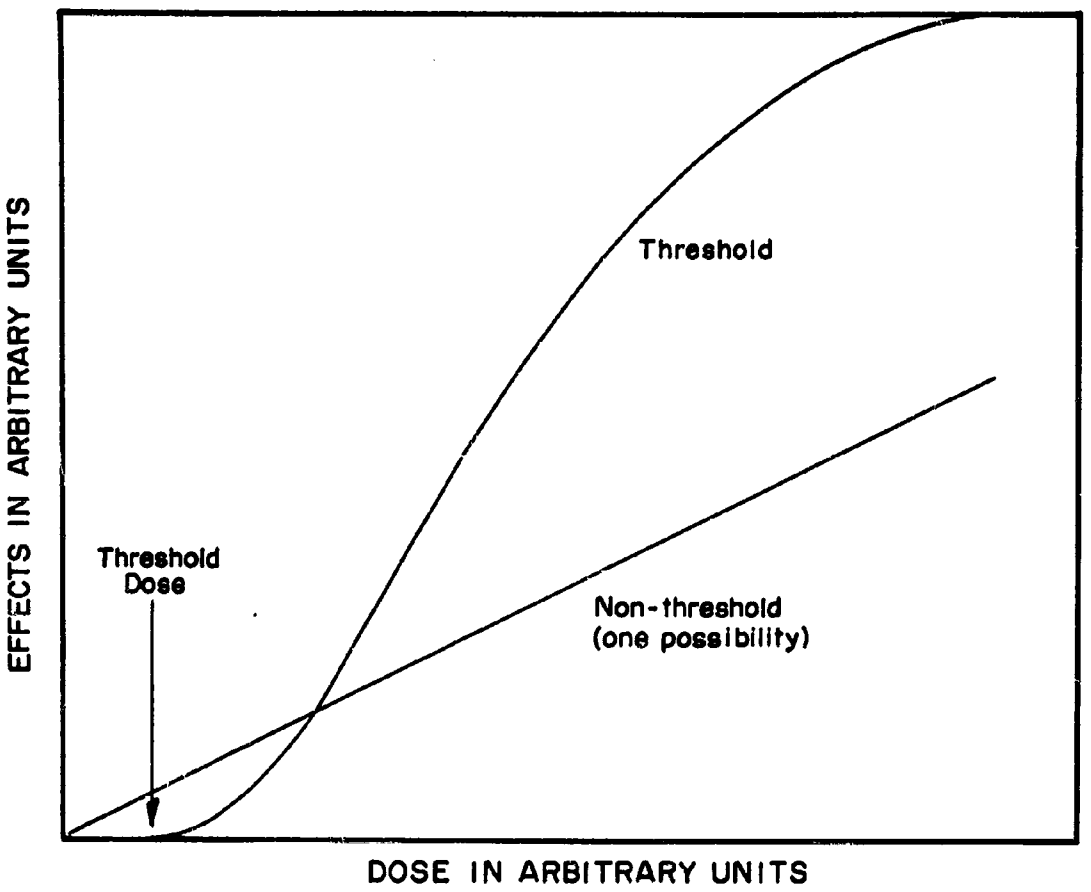


FIG. 9.7 REPRESENTATION OF TWO THEORIES ON SOMATIC EFFECTS OF IONIZING RADIATION

the problem of interpreting the data in the light of of biologic differences between man and the experimental animals.

One alternative approach to the study of the genetic effects of radiation is to attempt to determine the genetic changes which might occur in populations exposed to varying background radiation. Background radiation from natural sources is similar to fallout radiation in its physical nature and most likely in the quality of its biological effects. It consists of two types, terrestrial and extra-terrestrial in origin. Each type varies in intensity from place to place, the extra-terrestrial source varying with latitude and altitude, and the terrestrial source varying with the occurrence of the natural radioactive minerals in the soils. Whether background radiation is harmful or not is not yet thoroughly established, although some recent work seems to indicate a correlation between the number of congenital malformations occurring in certain areas and the level of background radiation[52]. Over a thirty year period the average background radiation deliver a dose of about 3 rems, while the fallout radiation in the north temperatc zones will deliver a dose of no more than a few percent of this amount. There are areas, however, such as in Kerala, India, where the background radiation can reach values as high as 24 rems over the thirty year period and it is here that fruitful studies of the genetic effects of radiation may take place.

Summarizing, therefore, it appears that there are many problems which need concentrated attention before a completely accurate estimate of radiation hazards can be made. While our knowledge of the levels of activity in the biosphere is fairly well established, there remains a need for fundamental research on the biological effects of this radiation. In view of these considerations it is clear that it is advantageous to maintain radiation levels as low as possible.

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CHAPTER 10

SUMMARY AND CONCLUSIONS

The High Altitude Sampling Program was initiated to permit the direct sampling of stratospheric radioactivity in order that the concentrations of fallout nuclides in stratospheric air might be measured accurately. A knowledge of these concentrations permits the calculation of the stratospheric burdens of strontium-90 and other potentially hazardous constituents of nuclear debris, and the determination of the mechanisms and rates of transfer of nuclear debris within the stratosphere and from the stratosphere to the troposphere.

Filter samples of stratospheric dust were collected by six Lockheed U-2 aircraft. The U-2 is a soaring aircraft which has a range of 3,000 miles and can operate at altitudes up to 70,000 feet. Throughout the program each aircraft was equipped with a duct sampler with the air intake located in the nose, and, during the last year of the program, with an additional duct sampler installed in a hatch on the underside of the fuselage. Both types of sampler were calibrated by means of in-flight measurements to permit accurate calculation of the volumes of air sampled. The filter medium used in both types of sampler was IPC 1478, a filter paper with high permeability, low ash content, low radioactivity content, and high retention for small particles filtered from high velocity air streams. It was developed by the Institute of Paper Chemistry in 1949 and has been widely used in programs of sampling of atmospheric radioactivity.

HASP sampling was limited to a single vertical-meridional plane because only six aircraft were available and it was necessary that frequent and regular observations be repeated in about the same localities over a long period of time to minimize sampling bias. It was assumed that zonal flow would

eventually transport all of the nuclear debris in the stratosphere through a meridional sampling plane. This plane was located at about 70° West (between 67° North and 7° South latitude) during November 1957 to July 1958, and at about 64° West (between 38° North and 57° South latitude) during September 1958 to August 1959. During September 1959 to May 1960 the sampling corridor ran from 80° West (at 11° North latitude) to 132° West (at 71° North) in order to solve certain operational problems. In May - June 1960 a brief resampling of the Southern Hemisphere was carried out. A total of 3,695 samples were collected and analyzed for strontium-90 and other nuclides during HASP. Of these, 2,956 were collected in the Northern Hemisphere and 739 in the Southern Hemisphere. Most of the filter samples were analyzed by means of carrier radiochemistry, with one sequential scheme of analysis used for strontium-90, cesium-137, barium-140, strontium-89, yttrium-91, zirconium-95 and cerium-144, with a second sequential scheme used for the analysis of rhodium-102, beryllium-7 and phosphorus-32, and with separate aliquots used for tungsten-185 or plutonium determinations.

The flight data for each HASP mission and the results of the radiochemical analysis of each sample have been presented in tabular form. These tables include, (1) the flight data needed to locate the collection site of each sample and to calculate the volume of air it represents, (2) the results of the routine analyses for the total beta activity and the concentrations of strontium-90, tungsten-185 and certain short-lived fission products, and (3) the results of the special analyses for cesium-137, plutonium, rhodium-102, etc. A cross section has been prepared for each mission showing the flight path followed during the collection of each sample and its relation to the tropopause structure.

The study of the HASP results and their comparison with data from other investigations of world-wide fallout have led to the formulation of several

conclusions concerning the behavior of radioactive fallout injected into the stratosphere by high yield nuclear weapons:

1. Radioactive debris introduced into the stratosphere by the nuclear weapon tests which were carried out between 1952 and the beginning of the moratorium on testing in November 1958 had still not become uniformly mixed throughout the stratosphere by mid-1960. Greater quantities of strontium-90 and of other fallout nuclides were present in the Northern Hemisphere than in the Southern Hemisphere, though the disparity was much less by 1960 than it had been during 1958 and 1959. Concentrations of strontium-90 and of other fallout nuclides normally increased with height above the tropopause but passed through a maximum value at some height, often still within the lower stratosphere, and then decreased with height at higher altitudes. The highest concentrations normally persisted at the latitude of the injection site, from which the debris mixed northward and southward, but stratospheric transfer processes could modify this situation.

2. The stratospheric burden of strontium-90, from all sources except the August 1958 rocket shots, Teak and Orange, was about 1.0 megacurie in mid-1958, but the burden rose sharply toward the end of 1958, with about 1.7 megacuries in the lower stratosphere below 40 millibars by December 1958. The burden was about 1.1 megacuries during 1959, with 0.6 megacurie below 40 millibars, and about 0.8 megacurie during 1960, with 0.5 megacurie below 40 millibars. During 1960 about 0.08 megacurie of strontium-90 from the rocket shots was also present in the lower stratosphere, giving a total burden of about 0.58 megacurie below 40 millibars.

3. The ratio of cesium-137 to strontium-90 in stratospheric debris was 1.7 ± 0.4 during 1958 to 1960. This ratio was virtually constant with latitude, altitude and time within the lower stratosphere.

4. The ratio of plutonium to strontium-90 in stratospheric debris was 0.017 ± 0.007 during 1958 to 1960. This ratio varied somewhat from place to place and from time to time depending on the source of the debris present.

5. The apparent residence half time of nuclear debris in the stratosphere, calculated from changes in the stratospheric strontium-90 burden, was about 10 months during 1958-1959 and about 18 months during 1959-1960. However, the stratospheric residence time of debris varied with both the latitude and altitude of injection. Debris stabilized in the lower stratosphere, below 40 millibars, exhibited a residence half time of about 6 months during 1958-1959 and about 12 months during 1959-1960. It appeared that debris injected into the

polar stratosphere by high yield Soviet tests had a mean stratospheric residence time (τ) of 4 to 6 months (varying somewhat with the time of injection) and that debris injected into the tropical stratosphere by United States and United Kingdom tests had mean stratospheric residence times which depended very much on the altitude of stabilization: from about 12 months, for debris which stabilized within 10,000 feet of the tropopause, to 5 years or more, for debris which stabilized above 100,000 feet.

6. The observed stratospheric distributions of tungsten-185, strontium-90 and other fission products and the changes which occurred in these distributions with time were consistent with the primary importance of turbulent diffusion for effecting the transfer of debris within the stratosphere. The concept of a meridional circulation of the stratosphere is difficult to maintain in the face of the persistence of the maximum concentrations of tungsten-185 in the lower tropical stratosphere for more than eighteen months following the injection of this nuclide, and its observed spread into the lower polar stratosphere along mixing surfaces which sloped toward the pole. If a meridional circulation does exist in the stratosphere, it is a second order effect at best as far as the transfer of radioactive debris in the stratosphere is concerned.

7. From the observed spread of tungsten-185 and of strontium-90 from specific shots it has been calculated that the meridional horizontal mixing coefficient, k_y, in the stratosphere is about 10^9 cm^2 sec^{-1}, and the vertical mixing coefficient, k_z, is about 10^3 cm^2 sec^{-1} or less in the tropical stratosphere and about 10^4 cm^2 sec^{-1} in the polar stratosphere.

8. HASP data have given direct evidence on the nature of mixing and transfer within the lower stratosphere and indirect evidence on the nature of these processes in the upper atmosphere. In the lower stratosphere, tropical air mixed into the polar stratosphere within layers which sloped toward the poles, as indicated by the behavior of stratospheric tungsten-185 and confirmed by the behavior within the stratosphere of strontium-90 and probably by that of ozone and water vapor. The behavior of stratospheric beryllium-7 and phosphorus-32 suggested that equatorward mixing of polar air may have taken place within the same layers. Large scale north-south meandering of streams of air was evident within the polar stratosphere, and this must have been a major factor in promoting meridional mixing. The involvement of tropical air within these meandering streams, especially during the winter season, probably produced the observed large fluctuations with time of concentrations of debris at specific points within the stratosphere at intermediate and high latitudes. Meridional mixing through 6,000 miles proceeded rapidly compared to vertical mixing through 8 miles.

9. Indirect evidence of mixing and transfer within the upper stratosphere was obtained by observation of the influx of Hardtack debris (apparently injected into the high tropical stratosphere by high yield Hardtack weapons) into the northern polar stratosphere during early 1959 and into the Southern Hemisphere during mid-1959, and by observation of debris from the rocket shots (injected into the mesosphere during Hardtack) into the northern polar stratosphere during early 1960 and into the southern polar stratosphere during mid-1960. These observations were compatible with the existence in the high stratosphere of "mixing layers" similar to those found in the lower stratosphere. The fact that debris from rocket shots, identified by its content of rhodium-102 and cerium-144, entered the lower stratosphere at high latitudes, in spite of its injection at low latitudes, indicated the more stable character of the upper tropical stratosphere compared to the upper polar stratosphere. Debris from these rocket shots appeared to display a mean residence time of over 5 years, though uncertainties in the initial Ce^{144}/Sr^{90} ratio in weapon fission products may have led us to an underestimate of the quantity of rocket shot debris in the lower stratosphere in 1960 and a resulting overestimate of the stratospheric residence time of debris from both surface bursts and the rocket shots.

10. The calculated rate of vertical mixing through the tropopause was considerably smaller than the observed fallout rate, and much of the loss of debris from the stratosphere must have occurred by some other mechanism, most likely by horizontal turbulent mixing and by organized meandering of air currents through the tropopause gap region. Evidence that meandering currents of air do transport parcels of air from the stratosphere to the troposphere and vice versa may be found in measurements of strontium-90 and of ozone in the stratosphere and troposphere.

11. Measurements of fallout at the earth's surface indicated that between 0.4 and 0.5 megacurie of strontium-90 fell out during 1955-1956 and during 1956-1957, about 0.9 megacurie fell out during 1957-1958, about 1.3 megacuries during 1958-1959, and about 0.6 megacurie during 1959-1960. The surface burden of strontium-90 was about 2.3 megacuries in July 1958, about 3.6 megacuries in July 1959, and about 4.2 megacuries in July 1960. These estimates have an uncertainty of $\pm$ 40%. In the absence of a renewal of weapon testing, fallout after July 1960 should result in a maximum surface burden of about 4.5 megacuries in 1962-1963, which should subsequently decrease almost with the half life of strontium-90.

12. In spite of the large amount of data obtained during HASP, some uncertainty remains as to the exact quantity of strontium-90 which was injected into the stratosphere during 1952-1958 and the exact amount which was still there

at any specific time between 1952 and the present. Mainly, the remaining controversy concerns debris injected at altitudes of 100,000 feet and above. Some disagreement still exists as to the mean stratospheric residence time of debris, though most authorities agree that residence time varies with latitude and altitude of injection, that polar injections exhibit residence times of a few months, and that tropical injections exhibit residence times between several months and a few years. Many authorities still assume that stratospheric transfer occurs mainly by means of a meridional circulation, though HASP tungsten-185 measurements and other data have convinced others that such transfer is essentially by turbulent diffusion. Though HASP data have added to the evidence for transfer of stratospheric debris to the troposphere by way of the tropopause gap region, it cannot be said that there is yet any general agreement as to the main mechanism by which fallout of debris from the stratosphere occurs.

13. The short mean stratospheric residence time of nuclear debris which is indicated by HASP data probably resulted in the rapid attainment of maximum concentrations of strontium-90 in human bone in 1959. If biologic uptake of strontium-90 varies more directly with the rate of fallout than with the cumulative surface deposit, concentrations in newly formed bone will decrease rapidly in future years, unless weapon testing in the atmosphere is resumed. The genetic dose rate from cesium-137 is only a small percentage of the MPD, and may be of less significance than the dose rate from carbon-14 when the infinite time dose to many generations from artificial carbon-14 is considered. The short stratospheric residence time of nuclear debris results in a greater potential hazard from short-lived gamma emitters than was once realized. In addition the radiation dose delivered to the lungs, lymph nodes and gonads by plutonium, which enters the body mainly by inhalation, appears to constitute about the same fraction of the MPD as does the dose delivered to the bone tissue by strontium-90. Evidently the irradiation of the human population by world-wide fallout from weapons tests performed before 1959 has not reached and will not reach levels which, by current standards, would be considered hazardous.